GREEN CAST DEMONSTRATION OF INNOVATIVE LIGHTWEIGHT CONSTRUCTION COMPONENTS MADE OF RECYCLED ASH FOR SUSTAINABLE BUILDINGS

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Abstract

The present article describes the main objectives and outcomes of the GREEN CAST project. This project aims at the demonstration of an innovative material that has been ideated and developed to be a green alternative to autoclaved aerated concrete (AAC). The present material is based on geopolymer technology and consists on fly ash activated in an alkaline solution in the presence of a foaming agent. The mechanical, thermal and acoustic characterization suggests that the properties of the developed material are similar to those of AAC. The life cycle analyses carried out for both materials reveal that the alternative technology has a lower environmental impact, associated to the use of the alkaline activator. The material in form of blocks was used to build a demonstrator and its behavior as thermal and acoustic insulating material was monitored and compared to an identical AAC made demonstrator. The similar performance of both materials reveals that GREEN CAST is a technical and environmentally viable alternative to AAC.

Keywords: *Lightweight, Fly ash, Autoclaved Aerated Concrete, Geopolymer.*

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1. INTRODUCTION

There is an increasing environmental need to develop new and more sustainable production technologies to utilize waste to substitute for raw materials in different industrial areas and reduce the CO_2 released to the atmosphere. In fact, the European Commission has committed to reduce the CO_2 emissions to 90´s levels by 2020. It is a major target to find alternative construction materials for the construction industry, because:

- The cement industry is the second one in CO_2 emissions to the atmosphere, at least 5% of total anthropogenic emissions (3.3 billion tons of cement are produced yearly in the world)
- The extremely large consumption of natural resources and generation of huge amounts of waste
- Millions of tons of waste potentially useful for the construction industry are generated in Europe yearly.

Every year, coal combustion fly ash is produced as a byproduct of combustion processes in thermal plants worldwide in large amounts. Although fly ash has not been classified as hazardous waste, it causes environmental concern due to the trace concentrations of heavy metals and crystalline silica that are present in their composition, limiting the amount that can be landfilled, or be in contact with water streams. On the other hand, fly ash can be classified as a pozzolanic material, which means that upon alkaline activation they harden exhibiting cementitious properties, and that is why they are mainly reused as a supplementary cementitious material (SCM) in cement and concrete composition.

Foamed, cellular or aerated autoclaved concrete (AAC) is defined as a cement-based material that contains at least 20% air in its structure. This ensures desirable properties such as low density and good thermal and acoustic insulation. The main drawback of conventional cellular concrete production is that has to be carried out in an autoclave process that involves high temperature and pressure.

Moreover, in the fabrication process of ordinary Portland cement (OPC), CO_2 is released into the atmosphere in the same quantity as material that is produced. This CO_2 is produced in the burning of fossil fuels and in the de-carbonation of limestone which is used as a raw material for the clinker. So far, in the preparation of cement based materials, fly ash has partially replaced commercial Portland cement in several types of OPC up to 35%.

By considering all the facts outlined above, the GREEN CAST project has emerged as a smart solution that proposes the full replacement of OPC by fly ash, in order to obtain non-structural lightweight precast construction elements with good insulation properties, which is additionally efficient from an environmental point of view.

GREEN CAST proposes to use fly ash as the main raw material, optimizing their management as waste, thus reducing their exposure to environment and therefore the associated risks related to their potential toxicity and environmental impact. Very importantly, compared to OPC, the CO_2 released into the atmosphere will be dramatically reduced, since neither fossil fuel nor carbonates are present in the GREEN CAST production process.

The main outcome of this research is the development and demonstration on an innovative construction product with good thermal, mechanical and acoustic properties, economically competitive and with minimized environmental footprint. The present manuscript relates the preparation and characterization of the material, as well as the demonstration step and the life cycle analysis, demonstrating the environmental benefits of the GREEN CAST technology with respect to its traditional competitors.

2. MATERIALS AND METHODS

The fly ash used in this project has been supplied by Cencatra, a waste management company located in Spain. Sodium hydroxide (50%) and hydrogen peroxide (33%) were supplied by Panreac. A commercial bubble stabilizer was used as received. All materials were used as received without any further purification. The fly ash composition was analyzed by X-ray fluorescence (XRF). This technique allows detecting chemical species and their content in a given sample. The particle size and distribution were analyzed by granulometry. A Malvern Mastersizer 2000 equipment was used. Measurements have been made after ultrasonic dispersion of samples by using water as dispersant.

The materials were homogeneously mixed with a mechanical stirrer in plastic recipients in the following way: first the sodium hydroxide solution was diluted and allowed to cool down at room temperature. Then, the bubble stabilizer (BS) was added to the solution and stirred. The fly ash was weighted and placed in the corresponding recipient, then homogenized mechanically. In each case the aqueous solution was added and mixed until complete homogenization. Finally the hydrogen peroxide solution was added to the mixture and mixed. The liquid mixtures were then immediately poured into the corresponding mold and covered with plastic film in order to prevent evaporation. The foaming process was allowed to take place prior to the curing stage. For the mechanical analyses, 16x4x4 cm specimens were prepared in metallic molds. The blocks were fabricated in PTFE coated wood molds (25x63x7 cm) with capacity for 5 independent blocks, **Figure 1**. All the samples were cured in laboratory furnaces at 80ºC for 16 hours. In order to analyze the foaming process of the material, a metallic mold with a transparent PMMA face was built.

Fig 1 Mixing process (left) and PTFE coated wood molds (right).

The density (ρ) of the GREEN CAST samples was analyzed according to the UNE-EN 772-13 standard. Six specimens were extracted from different parts of three blocks.

The compressive strength (M_C) was determined by following the standard UNE-EN 772-1. A constant load was applied at a constant rate of 0.1±0.05 MPa/s. The specimens were kept at a constant humidity of 6±2% prior to the tests. The dry density value is obtained from drying the specimens to constant mass.

The thermal resistance and conductivity was measured in a NETZSCH HFM 463 Lambda Heat Flow Meter, according to the UNE-EN 12664:2001 standard. The heat flow meter was used according to the norm ISO 8301:1991. In order to do that, 3 specimens were introduced horizontally in a heat flow meter, being the hot plate in the upper side. Said specimens were conditioned in a ventilation oven at 105ºC until constant mass is achieved, then coated with a waterproof cover which prevents them from moisture. The state temperature and the average temperature difference through the specimen were set at 10 and 15ºC, respectively.

The free-thaw resistance was investigated by following the UNE-EN 15304:2010 standard. Six cubic specimens (7x7x7 cm) were extracted from different parts of two blocks, saturated and kept for 48 hours in a polyethylene bag for stabilization and preserve moisture. The samples were placed in a chamber and exposed to 15 cycles of freeze (-15±2ºC) for 8 hours and thaw (20±5ºC) and humidity 95% for 8 hours. Both the mass and compressive strength loss of the specimens after this test were determined and compared to that of references taken from adjacent parts of the blocks.

In order to demonstrate the viability of the developed technology as constructive element, two mockups were built, one with the produced GREEN CAST blocks and an identical one built with commercial cement based AAC blocks. Both mockups were built in ACCIONA´s facilities in Alcobendas, Spain. The mockups consisted in metallic structures anchored to onsite casted concrete ground slabs and the two types of blocks were used to build the walls of each of the mockups.

For the construction of the reference mockup, Ytong blocks were used. The dimensions of the blocks were 25x63x7 cm and the density specified by the supplier 550 Kg/m^3.

Both mockups possessed one window oriented to the east and one door oriented to the north. In both cases generic aluminum/polyurethane sandwich panels were used as ceilings. The blocks were glued together with insulating/waterproof mortar. Both mockups were coated externally with such mortar, and finally, power supply was installed within them.

The acoustic insulation of the developed material was studied by in situ measurements of the following parameters:

-Airborne sound insulation of façades, according to the standard ISO 140-5:1998. For this experiment, an omnipower sound source generating a sound field with a continuous spectrum was placed outside the prototype at 7 m from the analyzed façade. Then, the average sound pressure level is measured in a point at 2 m from the façade and 3 different points inside the prototype. The airborne sound insulation index was calculated as the difference between pressure levels, inside and outside, standardized by reverberation time. The airborne sound insulation index was calculated as the difference between pressure levels, inside and outside, standardized by reverberation time.

-Measurement of the sound reduction index. In this case, an omnipower sound source generating a sound field with a continuous spectrum was installed inside the prototype. The average sound pressure level was measured in a point at 1 m from the façade and 3 different points inside the prototype. Background noise reduction inside the prototype was recorded too.

The sound measurements were carried out with the following equipment purchased to B&K: omnipower sound source 4292, audio power amplifier 2716, microphone 4189, hand held analyzer 2250 and sound level calibrator.

Fig 2 Measurement of airborne sound insulation of façades: sound source (left) and microphone (right).

In order to study thermal behavior of the real scale demo-buildings, a data acquisition system was installed in both the reference and GREENCAST mock-ups. An Agilent 34972A Data Acquisition Unit with a built-in 20-bit resolution digital multimeter was selected as autonomous data logging system, combined with a pair of Type T thermocouples (Copper-Constantan) for internal temperature measurement (sensitivity 40 µV/°C, nominal accuracy ±0.5°C). A Davis Instruments Vantage Pro2 weather station was used for external temperature monitoring. Thermal sensors were previously calibrated using a FLUKE 724 Multi Function Calibrator, finding differences not higher than 0.1°C.

In order to analyze the environmental impact of the developed technology, a comparison between the life cycle assessments, for both the GREEN CAST technology and generic AAC was carried out, according to the guidelines given in the ISO 14040-44 series, by using the SimaPro 7 software. For the study a cubic meter of product was defined as functional unit and an annual production of 50.000 m^3 for both types of materials was assumed. The environmental impacts of each life cycle stage have been calculated using the CML impact assessment method, which is a problem-oriented LCA method developed by the Institute of Environmental Sciences (CML) of Leiden University, the Netherlands. The data was directly obtained wherever possible in the field (primary data), when direct collection was not possible, derived data (secondary data) have been used, collecting on purpose data from literature or databases.

3. RESULTS AND DISCUSSION

The ash was characterized by XRF and SEM. As shown in **Table 1**, the ashes are mainly composed of SiO_2, Al_2O_3 and Fe_2O_3, with a low content in CaO, which corresponds to a F-class type of ash. The X-ray pattern, shown in Figure 1 reveals the mineralogical phases of this material: Quartz, Mullite, Hematite, magnetite, **Figure 3**.

Table 1 Fly ash composition analysis

	Al_2O_3	SiO_2	Fe_2O_3	TiO_2	CaO	MgO	Na_2O
%	27.5	44.0	17.3	0.9	4.2	0.9	0.18

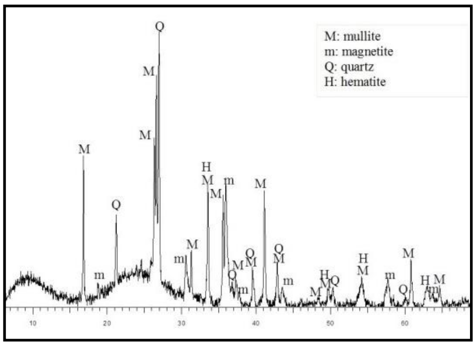

Fig 3 Mineralogical phases of the used fly ash.

Figure 4 shows the topology and internal microstructure of the fly ash. The particles are almost spherical and amorphous. Some spheres are hollow and others completely solid. The particles have a big range of sizes from 1.5 to 110 µm. The media size is 33.5 µm.

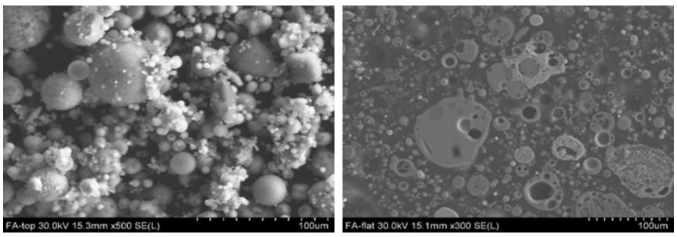

Fig 4 SEM micrographs of fly ash.

Previous research carried out showed that oxygen peroxide was the best performing foaming agent, and an optimization of the formulation proportions and the production process was carried out.

Formulations with different proportions of the components were prepared and tested. Considering the heterogeneous properties of the fly ash due to its waste origin, the formulation was optimized in a range of proportions. **Table 2** shows a range of accepted formulations.

Table 2 Range of weight percentages of each component in the mixture

Fly ash	NaOH	Water	H_2O_2	BS
67-71	6-8	20-25	0.4-1	0-0.15

The foaming process was investigated. A metallic mold with a transparent PMMA face was built and the successive mixtures were observed. It was found out that in the optimized range of proportions, the foaming process lasted for approximately half hour, and then it was allowed to stabilize for another one and a half hour prior to the curing step at 80ºC. **Figure 5** shows the foaming process over 30 minutes.

Fig 5 Foaming process at 0, 10 and 30 minutes.

By following this procedure, 400 blocks were produced, some of them were kept for characterization.

The material was prepared in form of blocks and their properties were measured. In **Figure 6** it is possible to appreciate a SEM micrograph of the porous structure of the material. The foaming agent added to the formulation generated a regular cellular structure that is the responsible for the low density of the material and its good insulating properties.

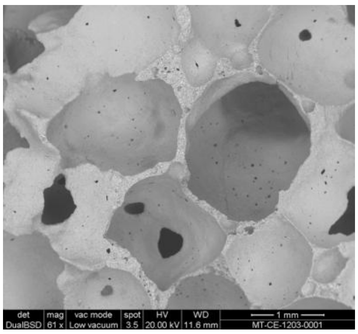

Fig 6 SEM micrograph of the GREEN CAST material (scale bar is 1 mm).

The main parameters of the samples were measured. The dry density was measured in 18 specimens extracted from 3 blocks. The measured density values are collected in **Table 3**. The average density was found to be 790±21 Kg/m^3.

Table 3 Density values for the analyzed specimens.

ρ (Kg/m^3)		
Block 1	**Block 2**	**Block 3**
774	811	801
764	803	813
767	784	798
762	795	810
765	799	805
766	803	812

Similarly, the compressive strength was measured by taking 6 specimens from 6 blocks; the values are shown in **Table 4**. The average compressive strength for the tested samples was 4.8±0.5 MPa.

Table 4 Compressive strength values for the analyzed specimens. B is for block.

M_C (MPa)					
B 1	**B 2**	**B 3**	**B 4**	**B 5**	**B 6**
4.1	4.7	4.2	5.1	6.0	5.9
5.1	5.5	4.7	4.7	4.4	5.0
4.2	5.4	4.0	4.7	4.7	4.8
4.7	5.3	5.0	4.5	3.9	5.1
4.9	5.0	4.7	4.7	5.7	5.1
4.4	4.0	4.9	4.4	4.0	4.3

The thermal properties of the materials were studied through the measurement of the thermal resistance and the thermal conductivity. Three specimens were tested and the results are shown in **Table 5**.

Table 5 Thermal resistance (R) and thermal conductivity (λ) of the analyzed samples

Sample	R (m^{2}K/W)	λ (W/mK)
1	0.3788	0.1880
2	0.3635	0.1955
3	0.3649	0.1648
Average	0.369±0.008	0.183±0.016

In the case of a cellular material, such as AAC, the thermal conductivity is directly related to its density, since the air present in the structure, lightens the material and reduces the thermal conductivity, therefore improving the insulating behavior. Conventional AAC presents typical λ values of 0.1-0.2 W/mK). The reference AAC used in this work, with a density of 550 Kg/m^3 presents a λ value of 0.16 W/mK according to the supplier. The slightly greater value of thermal conductivity obtained for the GREEN CAST material has been associated to its larger density.

The durability of the materials was analyzed by carrying out freeze/thaw cycles on the samples, measuring the mass and compressive resistance loss after said cycles. The temperature program consisted in cooling down to -15± 2ºC, keeping at that temperature for 8 hours and heating up to 20±5ºC, keeping at that temperature for 8 additional hours. **Table 6** summarizes the dry mass and compressive strength loss after 15 cycles compared to the references taken prior to the analysis.

Table 6 Mass and compressive strength loss after 15 freeze/thaw cycles.

		Mass loss (%)	M_C loss (%)
B1	**S1**	0.7	8.8
	S2	0.9	5.1
	S3	0.5	6.1
	S4	1.5	9.2
	S5	0.2	7.7
	S6	0.1	1.8
B2	**S1**	0.5	7.0
	S2	1.0	12.9
	S3	0.4	12.8
	S4	0.3	16.9
	S5	0.5	2.8
	S6	0.2	2.3

B is for block and S for specimen

For the tested samples, the average dry mass loss was 0.6±0.4 and the compressive strength loss was 7.0±4.0.

In order to demonstrate the constructive viability of the developed material, as well as its insulating behavior, a mockup was built with the GREEN CAST blocks and its performance was monitored. An identical mockup built with reference AAC blocks was built and monitored in the same way. **Figure 7** shows pictures of the constructive process of both prototypes, as well as a picture of both finished ones.

Fig 7 Mockups being built with GREEN CAST blocks (left) and Ytong ones (center). Both mockups finished (right).

The acoustic insulation of the developed material was analyzed by in situ measurements. This method allows characterizing the developed technology as part of a constructive system. In order to enable the comparison with a reference, the same measurements were carried out for the reference mockup.

The sound insulation index ($D_{2m,nT,Atr}$) is used to measure the level of sound insulation provided by a structure. This parameter can be expressed by a global value and by third octave bands. The results obtained for each of the façades of both mockups are shown in **Table 7**.

Table 7 Airborne sound insulation for the measured façades

	$D_{2m,nT,Atr}$ (dB)	
	GREEN CAST	**Reference**
North (door)	21.8	22.7
West	28.0	28.2
South	28.8	28.0
East (window)	25.1	26.4

In both prototypes the highest values for the sound insulation index were found in the West and South façades. This is because those walls do not contain elements with a low sound insulation such as windows. For this reason, these façades could be the most representative element of the prototype.

The sound reduction index of a material characterizes the sound insulation properties of such material or the constructive system itself. It was obtained from a modeling of the acoustic field. The results obtained for each of the façades is collected in **Table 8.**

Table 8 Sound reduction index (R_W) obtained for the measured façades.

	R_W (dB)	
	GREEN CAST	**Reference**
North (door)	21	23
West	32	30
South	33	31
East (window)	25	25

Identically, West and South façades presented the best R_W global values and in both cases both demonstrators showed very similar behavior.

Figure 8 depicts mockups internal temperature and external temperature from 7th August 2013 at 11 pm to 14th August 2013. Internal temperature for a given time is the result of averaging the output of two internal thermocouples placed at two different positions in the *xy* plane, keeping the height fixed at 1.5m

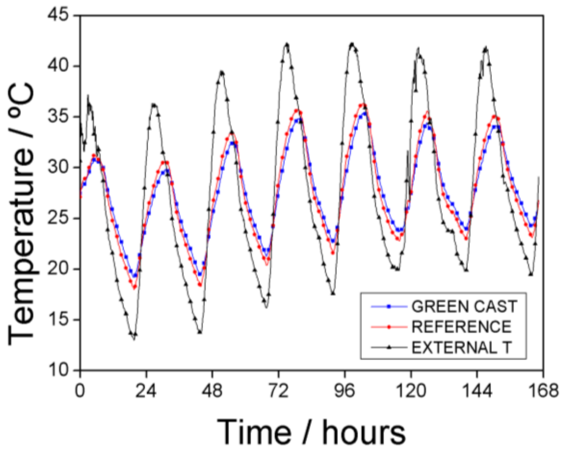

Fig 8 Mockups internal and external temperatures measured in a period of one week.

In the analyzed period, GREENCAST showed an attenuated thermal wave amplitude with respect to the reference one (3°C), maintaining the frequency. In both cases the temperature wave amplitude was found to be narrower than that for the measured external temperature, evidencing the insulating behavior of the materials. The insulating properties of both types of blocks showed to be practically identical, corroborating GREEN CAST as an alternative to current commercial solutions.

The ultimate aim of this work is to obtain an alternative to AAC with a lower carbon footprint. In order to quantify the environmental impact of the developed technology, LCA was performed for both technologies: GREEN CAST and the reference one. During the LCA study only the manufacturing phase was considered, performing a *cradle to gate* analysis, from raw materials acquisition through manufacture. The aim of a *cradle-to-gate* analysis is to assess potential environmental impacts associated with all the stages of manufacturing phase, without taking into account the subsequent phases crossing the *gate*. Fly ash is a by-product from coal combusted power therefore part of the environmental impacts of the electricity production should be allocated to the fly ash. In the case of by-products, two allocation procedures can be chosen: mass allocation or economic allocation. For this study the economic allocation procedure was used. According to Chen *et al*. [9] in the case of fly ash obtained in the production of power, the allocation by economic value for fly ash is 1% and 99% for electricity.
For this study, five impact categories were analyzed: global warming, ozone layer depletion, photochemical oxidation, acidification and eutrophication. **Table 9** summarizes the numerical results for each investigated impact category.

Table 9 Summary of impact categories values for generic AAC and GREEN CAST.

Impact Category	**Unit**	**AAC**	**GREEN CAST**
Global warming	Kg $CO_{2\,eq}$	182	80.5
Ozone layer depletion	Kg $CFC_{11\,eq}$	2 E-5	7.45E-6
Photochemical oxidation	Kg $C_2H_{2\,eq}$	0.0208	0.0176
Acidification	Kg $SO_{2\,eq}$	0.449	0.431
Eutrophication	Kg $PO_4^{3-}{}_{eq}$	0.037	0.03

As it has been evidenced, the potential impacts for acidification, eutrophication and photochemical oxidation categories are very close to those of AAC due to the emissions of sulphur and nitrogen oxides. It was found that the most significant parameter in both cases is the global warming, which is defined as the index used to translate the level of emissions of various gases into a common measurement to compare their contributions to the absorption by the atmosphere of infrared radiation. Although all the parameters were studied in detail, only the global warming has been included in this report due to its relevance in the present study. In the case of the traditional AAC, the major contributor to the global warming parameter is the cement, which acts as the binder and typically represents 20% of the weight of AAC. Individual contributions to this parameter for the GREEN CAST material are collected in **Table 10.**

Table 10 Contributions to global warming in the GREEN CAST technology.

Process Unit	**Quantity (Kg $CO_{2\,eq}$)**
Fly ash	0.08
H_2O_2	8.43
NaOH	44.8
Water	0.03
Raw material silos	0.1
Mixer	1.8
Blocks cutting	0.83
Oven	20.7
Blocks storage	0.83
Transport System	2.9
Total	80.5

Among all the contributions to the global warming, the most representative one is that from sodium hydroxide, followed by hydrogen peroxide.

In sight of the present analysis, it is possible to conclude that the developed alternative material possesses a lower environmental impact than conventional AAC associated to the absence of cement in its formulation.

4. CONCLUSIONS

A green alternative to AAC, in total absence of OPC, has been satisfactorily produced by the alkaline activation of fly ash in the presence of a foaming agent at relatively low temperatures and atmospheric pressure in a range of formulations. The physical properties have been measured under their corresponding international standards. The density, compressive modulus, thermal conductivity and durability properties were found to be very similar to traditional AAC. The viability of the GREEN CAST material as constructive element was demonstrated. Two identical mockups were built with each type of blocks and thermal measurements were carried out and compared to the exterior temperature for a period of one week, showing similar performance. Similarly, *in situ* sound insulation measures were carried out and the experimental data reveal the GREEN CAST material as good acoustic insulator as the commercially available AAC. A life cycle assessment was carried out for both types of materials, showing that the alternative material possesses a lower environmental impact and global warming index than AAC due to the absence of cement in its formulation.

It can be highlighted that a cellular material based on alkali activated fly ash, and in total absence of OPC, has been developed and compared to traditional AAC, showing a similar performance as constructive material and a lower environmental impact, standing out as a solid alternative to it.

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REFERENCES

[1]. Construction and Building Materials 2008, 22, 2393-2399.
[2]. ACI Materials Journal 2004, 6, 467-472.
[3] Resources, Conservation and Recycling 2011, 55, 669-685.
[4]. Journal of Hazardous Materials 2009, 172, 1049-1060.
[5] World of Coal Ash (WOCA) Conference, 2011
[6]. Cement and Concrete Research 2011, 41, 1244-1256.
[7]. Construction and Building Materials 2012, 28, 493-498
[8]. 2005 World of Coal Ash (WOCA), April 11-15, 2005, Lexington, Kentucky, USA.
[9]. Journal Resources, Conservation and Recycling 2010, 54 (12) 1231–1240

BIOGRAPHIES

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Dr. Giovanni Giodice received a University degree in Aerospace Engineering at Polytechnic University of Turin in 2002. Since 2003 he has been working as a researcher at the CETMA in the field of industrial design and scientific research. Main activities: Rapid Prototyping, Geographic Information System, Life Cycle Assessment, Environmental Analysis and Sustainable mobility.