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Combining LCT tools for the optimization of an industrial process: Material and energy flow analysis and best available techniques

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ABSTRACT

Life cycle thinking (LCT) is one of the philosophies that has recently appeared in the context of the sustainable development. Some of the already existing tools and methods, as well as some of the recently emerged ones, which seek to understand, interpret and design the life of a product, can be included into the scope of the LCT philosophy. That is the case of the material and energy flow analysis (MEFA), a tool derived from the industrial metabolism definition.

This paper proposes a methodology combining MEFA with another technique derived from sustainable development which also fits the LCT philosophy, the BAT (best available techniques) analysis. This methodology, applied to an industrial process, seeks to identify the so-called improvable flows by MEFA, so that the appropriate candidate BAT can be selected by BAT analysis. Material and energy inputs, outputs and internal flows are quantified, and sustainable solutions are provided on the basis of industrial metabolism.

The methodology has been applied to an exemplary roof tile manufacture plant for validation. 14 Improvable flows have been identified and 7 candidate BAT have been proposed aiming to reduce these flows.

The proposed methodology provides a way to detect improvable material or energy flows in a process and selects the most sustainable options to enhance them. Solutions are proposed for the detected improvable flows, taking into account their effectiveness on improving such flows.

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1. Introduction

For many years process design has focused on technical and economic factors, being the objective low investments and operational costs, high benefits and mechanized processes for reducing costs on manpower.

The World Commission on Environment and Development, known as the Brundtland Commission, was assembled by United Nations in 1983 in order to, among others, consider ways and means by which the international community could deal effectively with environmental concerns and help define a long term agenda for action during the coming decades [1]. The result was the publication in 1987 of the Report of the Brundtland Commission, "Our Common Future". It defined, for first time in history, "sustainable development", meaning a simultaneous economical, social and environmental growth to avoid deep ecology, conservationism or an excessive economical and social growth behind environmental consequences (Fig. 1). So, since then and progressively, modern plants need a different focus based on technical development aiming to integrate the three dimensions of sustainability.

Some philosophies emerged from the concept of sustainable development, being life cycle thinking (LCT) one of the most impactful ones. It can be understood in different ways. For the European Commission, LCT seeks to identify possible improvements to goods and services as lower environmental impacts and reduced use of resources across all life cycle stages [2]. On the other hand, for the United Nations LCT is about going beyond the traditional focus on production sites and manufacturing processes so that the environmental, social and economic impact of a product over its entire life cycle, including the consumption and end of use phase, is taken into account [3]. In spite of the focus considered, LCT is directly related to resources management under a life cycle point of view, involving not only environmental factors but the whole concept of sustainability.

LCT has been included in the European Union policies of the last decade, using as starting point the "Integrated Product Policy Communication" [4], as well as the "Thematic Strategies on the Sustainable Use of Natural Resources" [5] and the "Prevention and Recycling of Waste" [6]. All these strategies and methodologies are integrated in the "Sustainable Consumption and Production Action Plan" [7], which aims to reduce the overall environmental

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Fig. 1. The three dimensions of sustainability.

impact and consumption of resources associated with the life cycle of products.

According to Joint Research Centre [2], LCT drives towards sustainable consumption and production. Its life cycle perspective helps process designers to consider the resources consumed and the environmental impacts associated with the supply, use and end-of-life of products. Besides this new approach for process designing, LCT provides a series of tools that ease the complex task of integrating sustainability concepts in the process. That is the case of environmental management systems (EMS), design for environment (DfE), life cycle assessment (LCA) and material and energy flow analysis (MEFA) [8] among others. These are well known and globally applied tools, really useful when the project focuses on the environmental dimension of sustainability. Other methods, such as life cycle costing (LCC) or cost-benefit analysis (CBA) focus on the economics, whereas others, like social life cycle assessment (SLCA) or environmental impact assessment (EIA), combine environmental factors with social ones. This wide range of possibilities proves that the available tools are focused on specific dimensions of the sustainability [9], but none integrates the three of them. Therefore, there is a need to develop new methodologies under the LCT philosophy aiming the optimal integration of the social, economic and environmental aspects of the system through the application of tools that include the novel technical concepts.

Some authors are working on new methodologies based on the LCT philosophy combining known tools in such a way that one supplies the lacks of the other. That is the case of the *process design for sustainability*, a pioneer methodology proposed by Azapagic et al. [10] that enables the identification of relevant sustainability criteria systematically integrating sustainability into process design, or the most recent *life cycle assessment for sustainability* proposed by Heijungs et al. [11], which combines LCA and sustainability analysis to have a wide perspective of the analysed item. The present paper agrees with these works, as two known tools are combined into a single method to analyse and improve a process.

The goal of this paper is to develop a methodology under the LCT philosophy for an industrial process. This methodology is based on the combination into a single method of two known tools: MEFA and BAT (best available techniques) analysis. The first tool, MEFA, provides both qualitative and quantitative information about the improvable flows (IF) of the system, from environmental and eco-

nomic (indirectly) points of view. The second one, BAT analysis, uses technology to prevent and reduce the IF by applying BAT, defined by economic, environmental and social criteria. The method is applied to an exemplary roof tile manufacture plant for validation.

1.1. Material and energy flow analysis

MFA (material flow analysis) emerged from the industrial metabolism [12,13], defined by Ayres in 1988 as "the whole integrated collection of physical processes that convert raw materials and energy, plus labour, into finished products and wastes..." [14]. The concept "metabolism", both applied to biology and to society systems, appeared in the late 1860s, and it has evolved through history until the late 1960s, when it reappeared as a way to face the modern environmental concerns [13]. The publication of the book "Economics and the Environment: A materials Balance Approach" by Kneese et al. [15], set the basis of what would end up being MFA. It treated environmental and economic problems from a new perspective based on the utilization of mass and energy balances to evaluate the impact of the human activities over the environment. This flow perspective has been proved to be very useful when integrating society, technological change, and environmental effects [16] and, as Bringezu [17] stated some years later, it can be used to derive indicators for sustainability and analyse if sustainable industrial metabolism conditions are being met.

In spite of its origins, MEFA is typically used in accounting studies using accounting methodology and data [18], being the objective analysing the social metabolism of countries and regions [19]. Some examples are the works from Hendriks et al. [20], who applied MFA in a city and in a highly populated region to support policy making; Binder [21], who coupled social sciences modelling approaches and MFA to provide additional information to the management of material flows; Muñoz and Hubacek [22], who complemented MFA with a structural decomposition analysis and applied it to the Chilean economy; or Raugei and Ulgiati [23], who allocated appropriate percentages of LCA and MFA indicators to specific world regions, among others [24-26]. Other works complement this social approach with some economic analysis, aiming to understand socio-economic interactions [27,28]. Some examples are Kytzia et al. [29], who fed MFA with economic data to study causal relationships between economically motivated behaviour and resource consumption; Hawkins et al. [30], who combined MFA and economic input-output modelling to track economic transactions and material flows throughout the economy; or Krausmann et al. [31], who applied economy-wide MFA to quantify global materials extraction.

As an industrial metabolism based tool, MEFA has a huge potential when applied to industrial areas, where it has been successfully used to optimize material flows and waste streams in production processes [21]. However, there are not many examples of this approach. Some are the analysis by Sendra et al. [32] of an industrial park by MFA combined with water and energy indicators, the MFA of the US polyethylene terephthalate industry by Kuczenski and Geyer [33], or the application of MEFA to the transport and storage stage of a ceramics manufacture process by Torres et al. [8].

There is a lack of works that use MFA or MEFA to improve or evaluate industrial metabolisms. The original purpose of MEFA should be recovered to establish and improve symbiotic relationships in industrial systems or processes, combining it with other tools to consider, in an integrated manner, the three dimensions of sustainability.

1.2. BAT analysis

The European IPPC (Integrated Pollution Prevention and Control) Directive, in force since 1996 [34] and updated in 2008 [35],



Fig. 2. Flow sheet and environmental aspects of the selected process.

defines BAT as "the most effective and advanced stage in the development of activities and their methods of operation which indicate the practical suitability of particular techniques for providing in principle the basis for emission limit values (ELV) designed to prevent and, where that is not practicable, generally to reduce emissions and the impact on the environment as a whole".

BAT analysis, as proposed by Barros et al. [37], evaluates techniques candidate to be BAT following the method set by the EIPPCB (European IPPC Bureau) [36]. For each technique, this method considers environmental aspects, technical description, benefits, secondary effects, implementation, applicability and economical aspects, illustrated by example plants where the technique has already been applied [37,38]. BAT analysis implies a deep knowledge of the sector analysed, and an intensive study of the process itself.

As explained, literature provides some examples of how to combine MEFA with other tools so that the three dimensions of sustainability are taken into account. However, these tools only provide information about the system, but no solutions to improve it. A different option to complement MEFA is BAT analysis, which provides novel technical concepts to improve the sustainability of the process. In this work IF obtained by MEFA are used as starting point for the BAT analysis, obtaining solutions for a sustainable industrial process.

2. Methodology

The methods used in this work include the proposal of a new methodology and its validation in a real exemplary plant. Finally, the possibilities and applicability of the results obtained are discussed.

The proposed methodology integrates two tools: MEFA, applied to an industrial system to identify its IF; and BAT analysis to select of the most appropriate candidate techniques to enhance the process based on the identified IF. MEFA adapts the methodology proposed by Hendriks et al. [20], whereas BAT analysis uses the method adopted from the EIPPCB by Barros et al. [37].

The proposed methodology includes the following steps:

1. Definition of the system under study.

- Identification of the industrial plant.
- Qualitative analysis of the selected process: identification of its limits, definition and description of its stages, and analysis of inputs, outputs and internal flows. This analysis must be based on technical visits to the plant and on bibliographic review.
- 2. MEFA.
 - Data recompilation. Identification of the data needed to fill the model. The material and energy flows and stocks of the process can be determined by direct measurements (supported by technical visits to the industrial plant), best estimates, interviews, databases of environment protection agencies, scientific papers and so on.
 - Scenario modelling.
 - Software selection.
 - System modelling using the selected software. Scenario building creating networks and subnets. The model is fed with the data recompiled. Material and energy balances are performed on those processes where no data is available.
 - Model running and results of the MEFA. When it is complete, the model is run to obtain the results of the MEFA of the selected system.
- 3. Selection of the IF. The results obtained from the MEFA are analysed to identify the IF, meaning those inputs, outputs or internal flows that are remarkable in comparison with the other flows of the process.
- 4. BAT analysis
 - Recompilation and inventory of techniques candidate to be BAT for the selected industrial process.
 - Identification of the already implemented BAT. It gives an idea of the environmental performance of the plant and of the efficiency of the implemented techniques.
 - BAT selection according to IF. Candidate techniques from the inventory are selected for each of the identified IF.

3. Application of the methodology to an exemplary plant

The proposed methodology is applied, step-by-step, to a selected exemplary industrial plant.



Fig. 3. Qualitative analysis and flow allocation of the process.

3.1. Definition of the system under study

3.1.1. Identification of the industrial plant

The selected industrial plant, Cerámica Verea S.A., is a roof-tile manufacture plant located in Galicia, NW Spain, producing 111 millions of tiles per year (133.200t per day) (data from 2005). This installation is an exemplary industrial plant affected by the European Directive 2003/87/EC [39], concerning greenhouse gases emissions, and Directive 96/61/CE [34] concerning IPPC. It was the first existing Galician ceramics manufacture installation adapting to the requirements of IPPC Directive (transposed in 2002 to the Spanish legal system by Law 16/2002 [40]). The plant got the corresponding integrated environmental permit in 2005.

The plant performs a complete process regarding industrial metabolism. It includes physical, chemical and thermal processes requiring several raw materials and generating a variety of different waste streams.

3.1.2. Qualitative analysis of the selected industrial process

The productive process consists on five consecutive stages (Fig. 2) whose objective is producing roof-tiles from clay, which is the raw material of the process.

- Transport and storage of raw materials: Four types of clay from different sources are used. Clays 1, 2 and 3 are extracted from three different quarries next to the installation (no further than 7.5 km). Clay 4 is the clay mud resulting from quartz washing in the industrial and mineral rocks exploitation process carried out by a nearby installation. After extraction, clay is transported and stored in the plant. These activities mainly generate particles and exhaust gases associated to transport.

Raw materials preparation: Clay is loaded into the shredders and then into one of the four available dossing hoppers, which dose the appropriate combination of clays.

The four types of clay are mixed in the belt weigher, and the resulting mixture is preliminary milled in the roller mill. The clay mixture is finely grinded in the lamination and refining units, where the clay particles get the adequate size before storage.

The dust and particulate released to the floor, mostly by the lamination equipment, is weekly collected and fed to the storage stage (Fig. 3).

Shaping: It involves all the processes turning the milled clay mixture into raw tiles ready for thermal treatment.

The clay, previously loaded into a feed hopper, is added a CO_3Ba solution, which insolubilizes the SO_4Ca that clay may contain, avoiding possible efflorescence. Some clay is added a Mn_3O_4 solution to obtain graphite coloured tiles. After chemicals additions, the clay and its additives are mixed and added some water if necessary. The mixture is extruded through a mould, so the clay is shaped into a continuous piece of material which is cut. The resulting raw tiles are loaded into shelves that are conducted to the "thermal treatment" equipments. The green ware cuttings are fed back to the mixer so that no clay is wasted (Fig. 3).

Thermal treatment: The raw tiles are subjected to a thermal treatment integrated by three processes, namely drying, previous storage at the pre-kiln, and cooking (Fig. 4).

• *Drying*: The plant uses a counter current tunnel dryer divided into two sections, Dryer 1 and Dryer 2, where the raw tiles are set in wagons to be dried with pre-heated air from the kiln.

In Dryer 1 humidity is reduced. This dryer has two distinct zones with different air inputs. In zone 1 air is a mixture of fresh air and used air fed back from zone 1 itself. Zone 2 uses a mixture of air from zone 3 and fresh air.

Once tiles have gone across Dryer 2, their humidity is reduced. The counter current input air in Dryer 2 is a mixture of fresh air together with air from the cooling zone of the kiln. This dryer has a burner used to heat the air entering the dryer.

Tiles leaving the dryer go through an artificial vision system where their dimensions, weigh and structure are verified. The out-of-specifications tiles are removed and fed back to the milling stage so that they can be reused in the production process.

- *Pre-kiln*: the dried tiles are temporary kept in the pre-kiln to avoid humidifying from the ambient air. Pre-kiln uses air from the cooling zone of the kiln, which has been previously heated in a burner.
- *Cooking*: in the continuous tunnel kiln, tiles are cooked to get the appropriate properties by complex chemical reactions governed by the mineralogical, chemical and granulometric composition of the material [41]. At first tiles are slowly preheated. Then the temperature quickly rises, and the tiles get properly cooked. Finally they are cooled by cooling air that is later used in the dryer. The heat source is natural gas combusted in a burner. Broken cooked tiles are used as filler in the quarries.
- This stage generates emissions such as inorganic compounds, metals and VOC (volatile organic compounds) associated to chemical reactions and natural gas combustion. Besides, particulate and air are also released.



Fig. 4. Qualitative analysis and flow allocation of the "thermal treatment" stage.

 Post-processing: During post-processing the final product, tiles, are palletized before expedition. This stage involves activities such as packaging, palletizing and storage, which consume auxiliary materials (mainly packaging) and energy (both electricity and fuel) and release solid waste and exhaust gases (Fig. 3).

3.2. MEFA

The MEFA corresponding to the "Transport and storage of raw materials" stage has already been presented and evaluated in a previous work [8], so only the important results needed for the purposes of this work are presented here.

3.2.1. Data recompilation

All data used in this study was provided by Cerámica Verea S.A., being 2005 the time boundary [42]. Some data was measured in situ, other was obtained from tests at the laboratory and other was directly gotten from the on-line monitoring of the process. The data has been classified according to the process division shown in Fig. 2.

Tables 1 and 2 show the data recompiled concerning, respectively, electricity consumption and material flows. Though these tables show most of the information needed to feed the model, it is necessary to include some particularities about some stages:

- *Shaping*: some of cuttings from other stages of the process are fed back to this stage.
- Thermal treatment: the specifications for all the flows involved in this stage are displayed in Fig. 5. On the other hand, in the cooking process, tiles are at first slowly pre-heated to 650 °C. Then the temperature is quickly risen to 1054 °C, so that tiles get properly cooked.
- *Post-processing*: Tables 3 and 4 include the specific requirements of this stage.

3.2.2. Scenario modelling

3.2.2.1. Software selection. The software selected for this work is Umberto [43], developed by Ifu Institut für Umweltinformatik

Hamburg GmbH in cooperation with Ifeu-Institut für Energie-und Umweltforschung Heidelberg GmbH).

Umberto allows modelling, calculating and visualizing material and energy flow systems. The user can create individual projects and define several scenarios within a project. Material and energy flow networks in Umberto consist of these different types of elements [43]:

- Transition: It is a site in the flow network where materials and energy are transformed. They are represented by squares. A transition can be specified using the coefficients between input and output flows (linearly dependent) or using mathematical expressions (non-linear processes).
- Place: It is a site in the flow networks where material and energy are stored or distributed. They are represented by a circle. There are four different types of places:
 - Input: These places are the boundaries of the network or the part of the network to be examined with regard to the flows of material and energy entering the system.

Table 1

Electrical consumption of the stages of the process.

Stage	Process	Electricity ^b (kJ/kg)
Raw materials	Shredding and dossing	5.25
preparation ^a	Milling	6.51
	Primary lamination and refining	19.57
	Storage (transportation)	3.22
Shaping	Storage of clay in the hoppers	1.33
	Mixing	21.83
	Extrusion and cutting	67.63
	Shelves loading	1.02
Thermal treatment	Drying	107.45
	Pre-kiln	40.50
	Cooking	759.29
Post-processing	Storage (transportation)	3.5

^a The electricity to operate the conveyor belts is included in the electrical consumption of each engine.

^b All the electrical consumptions are referred to 1 kg of clay.

Table 2

Material flows and specifications of the process.

Stage	Material	Flow specifications		Rejects (%)	
Raw materials preparation ^a	Clay 1 Clay 2 Clay 3 Clay 4	24% 20% 8% 48%	50 t/hª	-	
Shaping	Clay mixture Mn ₃ O ₄ Ba CO ₃ Water	8.38 t/h 3% ^b 3‰ ^b 27.5% ^b	- - -		
Thermal treatment	Tiles	-		Drying Cooking	0.89 ^c 0.32 ^d
Post-processing	Pallets Band (11.6 mm) Carton Band (14.6 mm) Retractable plastic Wooden pallets ^e Labels	14,976 pallets 0.360 kg/pallet 0.323 kg/pallet 0.062 kg/pallet 0.500 kg/pallet 9.825 kg/pallet 0.0015 kg/pallet		- 4.0 0.0 2.6 4.0 -	

^a Total clay flow.

^b Weight content regarding clay flow.

^c To raw material preparation.

^d To quarries as filler material.

^e 16% Of the pallets are reused in the plant.



Fig. 5. Input data to model "thermal treatments".

- *Output*: They fix the boundaries of the system regarding the flows leaving the system.
- *Storage*: These elements are places where materials can be stored.
- *Connection*: These places can only distribute flows and cannot act as storage.

Furthermore, there are two special place types:

- *Input/output*: These places combine the function of input place and output place in one single place.
- *Port*: This element represents the link between two network layers: a net and its subnet.
- *Arrows*: These elements link places and transitions. They show the direction of materials and energy flows.

3.2.2.2. System modelling. The main network (Fig. 6), or 1st level network, of the model includes five different transitions, each corresponding to one of the five stages of the process (as shown in Fig. 2), connected by connection places. Each of these transitions includes a subnet (2nd level network), where all the correspond-

Table 3Pallet transportation (inside the plant).

	Transportation to storage area	Lorries loading
Diesel (L/h)	4.2	4.2
Load (t)	1.8	1.8
Worked hours (h/day)	5	4



Fig. 6. Process networks (1st level, 2nd level, 3rd level).

ing sub-stages identified in the qualitative analysis (Fig. 3) are modelled as transitions. Owing to its complexity, the transitions included in "thermal treatments" are modelled as subnets (3rd level network), corresponding to the qualitative analysis in Fig. 4.

Inputs, outputs and internal flows are places. Places representing inputs are raw materials (namely the four types of clay), secondary materials (water, BaCO₃, Mn₃O₄), operating supplies (electricity, diesel, natural gas, air) and auxiliary materials (packaging). Places representing outputs are atmospheric emissions, waste and rejected materials. Finally, input/output places are the recycling flows of clay and air.

Table 4

Expedition data for the finished product.

Average covered distance (km)		200
Lorry weigh (t)		15
Lorry load (t)		25
Road categories (%)	Highway Secondary road	25 55
	Urban road	20

The transitions involved in "raw materials preparation" and "shaping" subnets are linear. Therefore, coefficients have been determined for each input and output flow in all the transitions. These coefficients are calculated by simple material balances on the basis of 1 kg of clay mixture, using the data on Tables 1 and 2. The coefficients corresponding to each flow involved in these transitions are all displayed in Fig. 7. Flows are classified as inputs or outputs, indicating the materials involved, the places they come from or go to, and the variables assigned by Umberto for calculation.

The transitions of the 3rd level network are non-linear. Most of them represent thermal treatments that have been manually programmed considering mass balances on the basis of 1 kg of clay mixture. Data collected in Fig. 5 is also used, as well as information in Tables 1 and 2. Fig. 8 represents the allocation of input and output flows and the programmed mathematical expressions for some representative transitions.

For the drying subnet, transitions T1 (dryer – zone 1), T3 (dryer – zone 3), T4 (exchanger 1) and T6 (exchanger 3) are represented (Fig. 8). Some of the variables regarding air and clay humidity and temperatures are fixed on the basis of the data showed in Fig. 5. Others are calculated considering mass and enthalpy balances. T2

	T1. Shred	ding and dossing											
	11. Silled											_	
	Input / Output	Allocation Rules Cost Center Costs Co	st Drivers Constraints			_							
	Var Place	Material	Coefficient	B. Unit	DQ	^	Var	Place	Material	Coefficient	B. Unit DQ	J	
	▶ X00 P3	▲ Clay 1	0.21454	ł kg	٠		Y04	P5	▲ Clay mixture		1 kg 🤤		
	X01 P3	Clay 2	0.17878	8 kg	•								
	X02 P3	A Clay 3	0.52801	kg									
	X03 P3	A Electricity	5.29	k1									
	X05 P13	A Rejected dry tiles	0.00714	i ka									
	T3: Rolle	r mill											
п	Input / Output	Allocation Rules Cost Center Costs Co	st Drivers Constraints										
tio	Var Place	Material	Coefficient	B LInit	DO .	~	Var	Place	Material	Coefficient	B LINE DO	Т	
rat	X01 P2		6.51	k1	0	H	► Y00	P7	A Clay mixture	Coerricient	999999412 kg	-	
pa	X06 P5	Clay mixture	1	kg			Y01	P9	▲ Particle		5.88E-7 kg 🔮		
ore		1											
sl	14: Prima	ry famination and fem	ning										
ial	Input / Output	Allocation Rules Cost Center Costs Co	st Drivers Constraints										
er	Var Place	Material	Coefficient	B. Unit	DQ	^	Var	Place	Material	Coefficient	B. Unit DQ	Т	
lat	X00 P7	Clay mixture	1	kg	۲		▶ Y00	P8	Clay mixture		0.999735 kg 🔶		
L D	X01 P2	▲ Electricity	19.57	' kJ	•		Y01	P10	▲ Particle		0.000265 kg 🔶		
av	T5. Stora	Te										-	
R	15. 51014	ge		2								_	
	Input / Output	Allocation Rules Cost Center Costs Co	st Drivers Constraints										
	Var Place	Material	Coefficient	B. Unit	DQ	^	Var	Place	Material	Coefficient	B. Unit DQ		
	▶ X00 P8	Clay mixture	0.999734412	kg .	•		► Y00	P12	Clay mixture		1 kg 🗢	1	
	X01 P11	A Particle	0.000265588	i kg									
	102 72	Aclecticity	3.22	. N									
	T6: Partic	ulate collection for reu	itilization										
	Input (Output	Monstion Rules Cost Center Costs Co	t Drivers Constraints									-	
	Inpacy output y	alocadori Rules Cost Center Costs Cost	Configuration Constraints	0.11-3				DI	han to stat	C	D 11-7 DO	Т	
		A Particle	0.00221471	ka ka	00	-	Var ▶ voo	Place P11	A Particle	Coemcienc	1 kg	-	
	X01 P10	▲ Particle	0.99778529	kg		-	1.00	,			1 mg -		
							1					_	
	T1: Hopp	er											
	Input / Output	Allocation Rules Cost Center Costs Co	st Drivers Constraints	1									
	Var Diaca	Matarial	Coefficient	B LINK	Ino	~	Uar	Place	Material	Coefficient	R LINE DO	Т	
	▶ X00 P1	A Clay mixture	Coerricienc	1 ka	0	i	V01	Place P3	Clay mixture for graphite tiles	COOTTCIENC	0.03 kg		
	X01 P7	▲ Electricity	1.3	3 KJ	٠	_	V02	P12	Clay mixture for standard tiles		0.97 kg 🍳		
	T2. M. 20	N4 = 11/1/2 = 12											
	Input / Output	Allocation Rules Cost Center Costs Co	ost Drivers Constraints										
	Var Place	Material	Coefficient	B. Unit	DQ	^	Var	Place	Material	Coefficient	B. Unit DQ	T	
	▶ X00 P3	Clay mixture for graphite tiles	0.9	7 kg	٠		Y00	P4	▲ Clay and Mn3O4 mixture		1 kg 🗳]	
	X01 P2	▲ Mn304	0.0	3 kg	•								
	T3: BaCC	03 addition											
	Toput / Output		a patrona [caracterization]	1									
	Input / Output	Allocation Rules Cost Center Costs Co	Ta manual Constraints	In	la a l	120		let.	last set			т	
	Var Place	Material	Coefficient	B. Unit	DQ		Var	Place	Material	Coefficient	B. Unit DQ		
	X02 P4	Clay and Mn3O4 mixture	0.0307	8 kg	•		Y01	P5	▲ Clay and additives mixture for gra	P N	0.96913 kg		
	X03 P12	Clay mixture for standard tiles	0.9662	3 kg	٠		Γ						
-	T4. Minut	2				-						_	
ing	14. IVIIXei												
idt	Input / Output	Allocation Rules Cost Center Costs Co	ost Drivers Constraints										
Shi	Var Place	Material	Coefficient	B. Unit	DQ	^	Var	Place	Material	Coefficient	B. Unit DQ		
•1	▶ X00 P2	A Water	0.0059113	3 kg	•		▶ Y00	P6	Wet clay mixture for graphite tiles		0.03087 kg		
	X01 P7	Electricity A Clau and additions mixture for grad	21.8	3 kJ 2 ko			Y01	P6	Wet clay mixture for standard tile:	5	0.96913 kg 🔮		
	X02 P5	Clay and additives mixture for grap	0.3819231	z ky 5 ka									
	X04 P11	▲ Extruder graphite clay cuts	0.01852	2 kg	•								
	X05 P11	▲ Extruder standard clay cuts	0.58147	8 kg									
	T5. Darton	der/outtor											
	15. EXIL	del/cutter											
	Input / Output	Allocation Rules Cost Center Costs Co	ost Drivers Constraints										
	Var Place	Material	Coefficient	B. Unit	DQ	^	¥ar	Place	Material	Coefficient	B. Unit DQ	J	
	▶ X02 P7	▲ Electricity	67.6	3 kJ	•		▶ Y00	P8	▲ Shaped graphite tiles		0.03087 kg 🔮	1	
	XU3 P6	Wet clay mixture for graphite tiles Wet clay mixture for standard tiles	0.0308	7 Kg 3 kg			H YOI	P8	Shaped standard tiles		0.96913 kg 🍳		
		,	0.5091	- 1.4			1						
	T6: Shelv	ing											
	Input / Output	Allocation Rules Cost Center Costs Co	ost Drivers Constraints										
	Var Place	Material	Coefficient	B. Unit	DQ	^	Var	Place	Material	Coefficient	B. Unit DO	T	
	X00 P7	▲ Electricity	1.0	2 kJ	۲	Ī	► Y00	P9	▲ Shaped graphite tiles		0.012348 kg 🔶		
	X01 P8	Shaped graphite tiles	0.0308	7 kg	•		Y01	P9	A Shaped standard tiles		0.387652 kg		
	XU2 P8	Shaped standard tiles	0.9691	3 kg	•		¥02	P11 P11	A Extruder graphite clay cuts		0.018522 kg		
							H.00	,	a energia standara tidy tuts		SIGULTY BY	-	

Fig. 7. Transition specifications for "raw materials preparation" and "shaping" stages.



Fig. 8. Programmed mathematical expressions for some transitions included in the 3rd level networks.

(dryer – zone 2) and T5 (exchanger 2) have been modelled as T1 and T4, respectively, adapting the humidity and temperature parameters to the special requirements of these equipments, according to Fig. 5. The total electrical consumption of the dryer is programmed in T3. In this transition there have also been programmed the atmospheric emissions associated to the drying process, using the corresponding emission factors (Table 5). The rejected tiles factor (Table 2) is considered in T7 (stacking and artificial vision) to determine the amount of tiles rejected after drying.

The modelling for the pre-kiln subnet is similar the one presented for transitions T1 and T4 in the drying subnet, including also the calculation for the electrical consumption.

In the cooking subnet mass balances for air and clay are considered. The atmospheric emissions derived from chemical reactions

Table 5

Emission factors for combustion in the process.

Pollutant		Dryer – zone 3 (kg/t tiles) (data source [44])	Kiln (kg/t) (data source [45,46])	Industry machinery (g/kg fuel) (data source [47])
Inorganic compounds	NH ₃	_	_	0.007
C	CO ₂	35.500	61.00 ^a	_
	CO	0.155	$3.00 imes 10^{-2}$	15.80
	N ₂ O	-	-	1.30
	Chlorides (as Cl)	-	$4.00 imes 10^{-2}$	-
	Fluorides (as F)	-	$1.70 imes 10^{-1}$	-
	NO _x	0.049	$9.00 imes 10^{-2}$	48.80
	SO ₂	-	3.35×10^{-1}	-
Particles	PM ₁₀	1.150	4.35×10^{-1}	5.73
VOC	CH ₄	_	1.85×10^{-2}	0.17
	VOC	0.015	-	_
	NMVOC	-	-	7.08
	Benzo(a)pirene	-	-	30×10^{-6}
Metals	Arsenic (As)	-	1.55×10^{-5}	-
	Cadmium (Cd)	-	$0.75 imes 10^{-5}$	-
	Chromium (Cr)	-	2.55×10^{-5}	-
	Mercury (Hg)	-	3.75×10^{-6}	_
	Nickel (Ni)	-	$3.60 imes 10^{-5}$	-
	Lead (Pb)	-	0.75×10^{-4}	-

^a As kg/GJ.

are also calculated in T1 (pre-heating and firing), using emission factors (Table 5).

Finally post-processing subnet also includes non-linear transitions. T1 (packaging) considers simple mass balances to estimate the amount of packaging material used and rejected. T2 (transport to storage) calculates electrical consumption for the pallets electrically transported and the atmospheric emissions derived from transporting tiles using diesel vehicles. These emissions are calculated on the basis of annual diesel consumption, using data from Table 3, and considering the corresponding emission factors (Table 5). These calculations are the same for T3 (truck loading). T4 (delivery) uses a specific module library included in Umberto, which describes transport of goods by truck and considers travel distance, load, vehicle type and road type to calculate emissions corresponding to fuel combustion.

3.2.2.3. Model running and results of the MEFA. The model has been executed considering the raw clay input during one year, 2005. Table 6 shows the results of the global material and energy balance for the whole process. Materials and energy have been classified as inputs and outputs. Fig. 9 represents the most relevant Sankey diagrams obtained for the process. Umberto uses them to display materials, energy and costs flows.

Results obtained for the storage stage [8] concluded that transport was the most important source of pollution, regarding atmospheric emissions. This stage is a great fuel consumer and consequently a great exhaust gases releaser, being CO₂ the main pollutant. Another important environmental impact linked with transport is particulate matter.

The most remarkable point about "raw material preparation" is the electricity consumption. The processes involved in this stage require approximately 2.40×10^9 kJ of electrical energy, most of it consumed by the primary lamination and refining processes, though this consumption is not that relevant if compared with other stages.

The electrical consumption in "shaping" is 16.14×10^9 kJ, mostly demanded by the mixer and the extruder/cutter engines. However, the most relevant issue about "shaping" is not its electrical demand but its clay cuts flow. Clay cuts from shelving are totally fed back to the mixer, so that they are reused in the process. As a result of

this technique, no material is rejected and therefore no waste is generated.

Thermal treatments differ from the already mentioned stages as, unlike those ones, they not only demand electricity but also fuel, natural gas in this case. Natural gas consumption affects both the inputs and the outputs, as its combustion, besides heating, generates emissions. Most of these emissions are CO₂, as 11,627.12 t are released, mainly from the cooking sub-stage. However, it is not only CO₂ what is released because of the natural gas combustion but also other pollutants such as CO, NO_x, PM₁₀ or VOC. Non-CO₂ atmospheric pollutants become especially relevant on the cooking subnet, as pollutants resulting from the natural gas combustion get mixed with other compounds, such as metals, CH₄, chlorides and fluorides released as a consequence of the chemical reactions that take place during cooking, according to the reference literature [48].

Thermal treatments are typically the most polluting stage in the ceramics manufacture processing. However, as the considered plant uses a low-carbon fuel, natural gas, atmospheric emissions (mainly CO_2) are much lower than the ones expected.

The other relevant mass flows associated to the thermal treatments are the two rejected tiles flows, one corresponding to the defective dry tiles from the dryer and other to the broken cooked tiles from the kiln. Almost all the dry tiles are fed back to the "raw materials preparation" stage, in order to be milled and reused, whereas the cooked tiles are a residual flow which has to be conveniently managed.

Thermal treatments are highly energy demanding processes; in fact they consume 72.41% of the total amount of electricity required by the plant. Among thermal treatments, cooking is the one requiring more energy, as it demands 83.50% of the electricity provided to the stage and 84.96% of the total natural gas fed to heat the air streams. The cooking kiln needs an important fuel supply, natural gas in this case, in order to heat air to the high temperatures required to cook the tiles. Air inputs come both from the outside and from the cooling pits, whereas air outputs are air from the cooling zone which is used as pre-heating air in the dryer and the pre-kiln, as well as excess air that is released.

Regarding "post-processing", the most relevant fact about it is its fuel consumption, though irrelevant if compared with the one obtained for the "transport and storage of raw materials" stage [8]. Consequently CO_2 is emitted.

Table 6 Global balance for the roof tile manufacturing process.

	Item							Quantity	Unit
					Clay 1		14,921.00	t	
	D			(Clay 2	Moisture	20	12,434.00	t
	Rav	v mate	riais	(wet basis)	Clay 3	content (%)	35	36,729.00	t
					Clay 4		20	4974.00	t
					Water			3689.43	t
	500	ondom	mot	oriala	CO ₃ Ba		208,987.91	kg	
	300	onuary	mat	citais	Mn ₃ O ₄			64,541.62	kg
					Air			1,638,092.58	t
uts					Carton			17,457.27	kg
Inp					Labels			810.71	kg
					Band (11.6	mm)		19,457.02	kg
	Aux	kiliary	mate	rials	Band (14.6	mm)		3350.93	kg
					Wooden pa	llets		446,052.21	kg
					Re-usable p	allets		84,962.33	kg
					Retractable	plastic		27,023.64	kg
	_				Fuel oil			26,781.80	t
	Ene	ergy			Natural gas			3036.00	t
					Electricity			68.95×10^{9}	kJ
			Am	monia				535.07	kg
		~	Car	bon dioxide			99,849.36	t	
		rganic	Car	bon monoxide	176,348.98	kg			
			Din	itrogen monoxide	8922.85	kg			
		lmc	Chl	linondes Nuerides				2283.71	kg
		C I	Flue	orides	9820.36	Kg			
			NU.	X nhun diaxida			930,141.92	kg	
		Dorti	Sul					42,448.80	кg t
		1 4110	Mat	thane				3061.28	ι ka
	air		IVIC	Halogenated	Aromatia PCDD PCDE			1.60×10^{-6}	kg
	s to	0	U	Aldeled	Alomatic	Mathedana and	1.	1.00 × 10	1- r
	ons	ŏ	2	Aldenydes	HC	Niethylene oxi	de	68//.//	Kg
	issi	~	ž	Aromatic	HC	Benzene		1613.30	kg
uts	Em		z	PAH		Benzo(a)pyren	e	0.21	Kg
utp				Unspecified				/6,350.16	Kg
ō					Arsenic			0.85	Kg
					Charmium			0.41	kg
		Meta	ls		Moroury			0.21	kg
					Nickel			1.08	kg
					Lead			1.90	kg
					Purge from	drier		172 156 31	к <u>g</u> t
		Air			Air from dr	ier and pre-kiln		981 059 78	t t
	AII				Air from kiln			473 045 48	t
		Air Irom Kiln				111		778.28	ι kσ
	te	Band	(14)	6 mm)				87.12	kø
	Vas	Retra	ctabl	e plastic				1080.95	kg
		Rejec	ted o	cooked tiles				175.611.15	kg
	-				Graphite til	es pallets		1706.65	t
	Products				Normal tile	s pallets	53,595.34	t	

3.3. Selection of IF

The MEFA results have proved that "transport and storage of raw materials" and "Thermal treatments" are the most likely to be improved stages, because of their high resources consumptions and pollutants emissions. Both stages demand important amounts of combustible, meaning fuel and natural gas, depending on the stage considered. Furthermore they release relatively high amounts of atmospheric pollutants, mainly exhaust gases that, though they do not exceed the ELV fixed in the environmental permit [49], they are improvable.

Table 7 shows the IF identified after evaluating the results of the MEFA. For each stage, the quantitatively relevant flows (Fig. 9) have been selected as improvable. Therefore, 3 IF have been selected for both "transport and storage of raw materials" and "post-processing" stages. Only 1 IF has been identified for stages "raw materials preparation" and "shaping". Finally, 6 IF have been identified for "Thermal treatments".

Concerning their type, there have been identified seven inputs, all of them associated with energy, and seven outputs, all of them concerning waste streams.

3.4. BAT analysis

3.4.1. Recompilation and inventory of techniques candidate to be BAT

Barros et al. proposed, in 2007, a procedure to implement the requirements of the IPPC Directive to the heavy ceramic indus-



Fig. 9. Sankey diagrams.

Improvable flows identified for the process.				
Stage	Improvable flows			
	Inputs	Outpu		

	Inputs	Outputs
Transport and storage of raw materials	Diesel fuel	Exhaust gases Particulate emission
Raw materials preparation	Electricity	
Shaping	Electricity	
Thermal treatment	Natural gas Electricity	Rejected cooked tiles Particulate CO ₂ Fluorine and other atmospheric emissions
Post-processing	Fuel Electricity	Exhaust gases

Table 7

try in Galicia (NW Spain) [37]. In that work they presented a deep analysis of the sector in the region, identifying 21 ceramic manufacture plants affected by the Directive in Galicia. They also described in detail the generic typical process performed by the sector, which can be conveniently adapted to any of the integrating plants. Regarding the process, associated consumptions and emissions were identified, highlighting the special relevance of atmospheric emissions, mostly particulates and exhaust gases from thermal treatments. Finally, an inventory of techniques candidate to be BAT for the heavy ceramics industry was developed, following a specific method adapted from the EIPPCB. As a result, 37 techniques, classified attending to the process stage where they should be applied, were gathered together and complemented by a series of best environmental practices, both generic and specific.

3.4.2. Identification of the already implemented BAT

Cerámica Verea S.A. has already implemented four BAT, which has directly affected the results obtained for the MEFA. The already implemented techniques are:

- Installation of bulk storage areas for dusty materials enclosed with walling. It affects to "transport and storage of raw materials", "raw materials preparation" and "shaping" stages, reducing particulate and dust emissions. The effectiveness of this technique is quite good, as nor dust neither particulates have been quantified in "raw materials preparation" and "shaping" stages (see Fig. 9)
- Reprocessing of green and unfired wares. This technique, implemented in "raw materials preparation" and "shaping" stages, aims to benefit the residual usable ceramic. In the case of "raw materials preparation", rejected dry tiles are fed back to the process from the drying stage, whereas in "shaping" is green ware, as extruder clay cuts, what is fed back to the mixer (Fig. 9).
- Optimizing cooking and drying processes. Thermal treatments have been optimized by recovering heat from flue gases from the kiln and feeding it back to dryer. It is the best option for minimising energy consumption in such an energy demanding process
- Utilization of low ash fuels such as natural gas. Due to the typical relatively high emissions rate from firing, natural gas is used as fuel in the thermal treatments stage. In spite of being the most energy consuming stage, it is not the one releasing more atmospheric pollutants

3.4.3. BAT selection according to IF

Candidate techniques are proposed for those IF likely to be enhanced, as indicated in Table 7.

3.4.3.1. Transport and storage of raw materials.

- To enhance the IF "diesel fuel" *limiting vehicles speed* and *minimising transport distances* is proposed. Both measures affect diesel fuel consumption and exhaust gases emissions as well.
- The IF "exhaust gases" can be reduced by *limiting vehicles speed* and *minimising transport distances*. The last measure effectively reduces exhaust gases emissions as fuel consumption, and thereby combustion gases, directly depends on the travelled distance. Furthermore, the most consumed clay is Clay 4 (Table 2), the one collected from the furthest quarry, which is 15 km away from the plant, so encouraging the usage of the other clays could contribute to reduce two IF, namely "diesel fuel" and "exhaust gases".
- The techniques proposed to reduce the IF "particulate" are *limiting vehicles speed, minimising transport distances* and *circulating through paved roads*, which have been reported to reduce dust and particulate emissions [50].

3.4.3.2. Raw materials preparation.

- To enhance the IF "electricity", *energetic optimization* is proposed. This technique involves programming and using equipment only when necessary and in the most appropriate conditions.

3.4.3.3. Shaping.

- The IF "electricity" can also be reduced by *energetic optimization*, the same technique as described for "raw materials preparation".

3.4.3.4. Thermal treatment.

- The IF "natural gas" cannot be reduced, as it is itself a BAT, and it is also affected by another implemented BAT, *optimizing cooking and drying processes*. In fact, energy consumption could have been even greater in case of not re-using hot air in the process.
- As in the case of "natural gas", the IF "electricity" cannot be improved as energy consumption has already been optimized for this stage.
- The IF "rejected cooked tiles" is another IF that cannot be further improved. After cooking, tiles cannot be fed back to the process, so there is no chance to recover them. Besides there are no techniques that can effectively reduce the amount of tiles broken while cooking.
- The IF "particulate" can be highly reduced by the implementation of either *bag filters with injection of NaHCO*₃ or *electrostatic precipitators with injection of NaHCO*₃. Both techniques are able to reduce even more than 99% dust and acid gases emissions [41]. Other advantages are that, besides requiring some extra energy supply, they do not consume many raw materials and they barely release any waste. Moreover investment and operational costs are not too high [41], so the installation of such equipments can be easily afforded by the company.
- The IF "CO₂" cannot be further improved, as it is quite lower than the value expected for such a process. CO₂ emissions have been highly reduced, regarding typical ceramics manufacturing process, thanks to the already implemented technique *utilization of low ash fuels such as natural gas.*
- The IF "fluorine and other atmospheric emissions" also benefit from the techniques *bag filters with injection of NaHCO*₃ and *electrostatic precipitators with injection of NaHCO*₃, as they not only reduce particulate emissions but also sulphur oxides, fluorine and chlorine components [48].

Among the atmospheric pollutants included in this IF, fluorine is a special case. Though fluorine emissions do not exceed the ELV fixed by the competent authority [49] (and they are not even mentioned in the environmental permit), they exceed the BAT-AEL (BAT associated emission limits) proposed by the European Commission [48]. Besides the already mentioned techniques, this IF could be also reduced by implementing one of the multiple *techniques for reducing inorganic compounds*. However, these techniques, although effective, are not really applicable, as they demand chemical compounds and great amounts of water that has to be treated as waste water. Besides, economical investments are considerably greater than in the case of bag filters of electrostatic precipitators.

3.4.3.5. Post-processing.

- The IF "diesel fuel" can be enhanced by limiting vehicles speed and/or minimising transport distances.
- There are no candidate techniques that can effectively reduce the IF "electricity" in the "post-processing" stage.

- The IF "exhaust gases" can be reduced by the techniques *limiting* vehicles speed and minimising transport distances.

4. Results and discussion

The application of the proposed methodology to an industrial productive process has led to the identification of its IF by MEFA, and the selection of the most appropriate candidate BAT to enhance them.

Modelling the process and applying MEFA has enabled to the identification of the IF. 14 IF (7 inputs and 7 outputs, concerning both materials and energy) were identified. Six IF concern "thermal treatments" stage, which has been proved to be the most likely to be improved stage, whereas the stages involving transport, namely "transport and storage of raw materials" and "post-processing", have 3 IF each. The other 2 IF correspond to "raw materials preparation" and "shaping". All the input IF are related to energy, as they correspond to electricity or fuel flows. On the other hand, almost all output IF are atmospheric emissions but one, which corresponds to rejected cooked tiles.

The application of BAT analysis has led to the identification of 4 techniques that have already been successfully implemented. According to the identified IF, 7 candidate BAT directly affecting 9 IF have been proposed.

The already implemented techniques highly contribute to the exemplary environmental performance of the plant. In fact, for two of the stages of the process there have not been detected particle flows or rejected clay flows. In addition, a low ash fuel, natural gas, is used in thermal treatments. The optimized configuration of this stage is also a BAT. Consequently both fuel consumption and CO_2 emissions are much lower than those expected for a typical process.

On the other hand, among the proposed techniques *circulating through paved roads, limiting vehicles speed* or *minimising transport distances*, are really easy to implement and do not involve any additional cost for the plant. They can considerably reduce the IF particles, exhaust gases and diesel fuel used involved in "transport and storage of raw materials" and "post-processing". *Energetic optimization* of the equipments involved in "raw materials preparation" and "shaping" is also quite easy and cheap to implement. It includes good practices such as minimising pumping distances, adjusting the operating conditions to the needs of the process, controlling electricity consumption, or automating the process, among others. All these practices could mean an important reduction of the IF electricity.

The techniques proposed to reduce atmospheric emissions, namely bag filters with injection of NaHCO₃ or electrostatic precipitators with injection of NaHCO₃, and techniques for reducing inorganic compounds, are more difficult to implement. They involve economical costs, as equipment has to be acquired, chemicals are needed and waste streams are generated. However, the good performance data reported for these techniques justify their application, as IF regarding particulate and atmospheric emissions (especially HF) can be highly enhanced. The best option will be selecting either bag filters with injection of NaHCO₃ or electrostatic precipitators with injection of NaHCO₃. Besides reducing particulate emissions, these techniques are very effective to reduce acid gases emissions, such as HF, making it possible to meet the BAT-AEL 10 mg HF/Nm³.

Even when applied to such an exemplary plant as the considered one, the methodology provides important information about the IF and a list of candidate BAT with a technical sheet for each technique. These results could be used by decision-makers to select and implement the appropriate modifications to enhance the process under study, based on industrial metabolism and sustainable criteria.

5. Conclusions

This paper presents a new methodology intended to apply the LCT philosophy into an industrial production process. The method combines two known tools to identify the IF of the process and to propose the most appropriate techniques (concerning sustainability) to improve its industrial metabolism. The method has been validated in an exemplary ceramics manufacture plant. 14 Improvable flows have been identified for the process, six of them related to the "thermal treatments" stage. Consequently 7 candidate BAT have been proposed aiming to reduce these flows.

The combination of MEFA and BAT analysis has turned out to be a good option for process evaluation considering sustainability criteria. The application of the methodology to an exemplary process shows that MEFA gives deep information about the environmental aspects of the process to identify the IF, while BAT analysis helps taking into account not only environmental factors, but also economic aspects that may affect the selection of techniques. It is shown that even a plant with such an exemplary environmental performance as the analysed one can be improved by the implementation of the selected modifications.

The proposed methodology provides a way to detect improvable material or energy flows in a process, whether they are inputs, outputs or internal flows, and selects the most sustainable options to enhance them. Solutions are proposed for the detected IF, and their effectiveness on improving them is roughly considered, though not specific and quantitative data about these improvements is provided.

These results are very useful to enhance the process regarding resources consumption, environmental performance and, at lower level, economical aspects related to the habitual operation of the analysed process.

An interesting continuation for this work will be the implementation of the proposed techniques in the selected plant. This task is not easy to accomplish with, as it will require a strong implication of the plant, with important inversion and long period of time to fully implant and implement the techniques, and to analyse their effectiveness.

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