

Integrated pollution prevention and control for heavy ceramic industry in Galicia (NW Spain)

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Abstract

The heavy ceramic industry (building materials and refractory products manufacture) is an important source of pollutants to the environment. For this reason these industrial sub-sectors are included in prevention and control pollution policies, specifically those of the European Union. The IPPC Directive pays particular attention to the mineral industries, not least to the ceramic industry (epigraph 3.5, Annex I). In this paper, a methodology which is being applied to support IPPC installations and the competent administrative authority in Galicia (NW Spain) is presented. For that, the Galician heavy ceramic industry is analysed, as also are the ways to study the Best Available Techniques (BAT) with a view to establishing the emission limit values (ELV) for each specific case. Hence, a technological state of the art has been carried out for both sub-sectors, from the point of view of implementation of the IPPC in Galicia. Following this, the processes are described briefly and an analysis of the consumption and emission levels of the main pollutants is made. An inventory that includes the best environmental practices and the preventive and abatement candidate techniques as BAT was elaborated for both considered sub-sectors. An information data sheet for each candidate BAT is presented as a method to help both the industries and the competent authority to identify a candidate technique of the inventory as BAT. Three illustrative examples of the application of this procedure are presented for different emissions to environmental media for Galician installations.

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1. Galician affectation under the IPPC transposition in Spain

On 24th September 1996 the Council of the European Community issued the Directive 96/61/EC, the *IPPC Directive*, concerning integrated pollution prevention and control [1]. The IPPC Directive is one of the most ambitious legal measures that the European Union (EU) has initiated with a view to applying the prevention principle for industrial activities.

This integrated approach means that firstly all productive process stages are considered, secondly that a relationship between the quantity of the polluting emissions and the features of the receiving medium is established, and the polluting transference from one medium to another (air, water and land) is also considered. The IPPC aims to achieve the integrated prevention and reduction of environmental pollution emitted by those industrial

installations with a higher potential of emissions to the environment, enumerated in the Annex I of the aforesaid regulation.

The priority of the IPPC is to reach a high level of protection of the environment as a whole. Industrial pollution must be minimised at source by applying *Best Available Techniques* (BAT) to the productive processes considered. Likewise, *Emission Limit Values* (ELV) based on BAT must also be established for the process [2,3]. Both, BAT and ELV must be checked and periodically updated so that the latest technical developments can be taken into account.

The transposition of the Directive to the Spanish national legal system was made by the Law 16/2002, on 1st July, which concerns the integrated pollution prevention and control (*Ley 16/2002, de 1 de Julio, de prevención y control integrados de la contaminación*) [4], which due to its basic nature is a legally binding environmental rule for the whole country. Integrated pollution prevention and control has given rise to a new kind of *environmental permit*, which replaces and draws together a dispersed group of environmental permits required so far. This new scheme allows to establish an administrative simplification,

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organising a complex administrative procedure which integrates all the existing environmental permits related to production and waste management. Incineration permits and those of installations that discharge to continental waters and from land to sea are also included, as well as other requirements of an environmental nature contained in the sectorial legislation.

As the IPPC Directive states, the ELV for pollutants should be based on BAT without prescribing the use of any specific technology, but taking into account the technical characteristics of the concerned installation, its geographical location and the local environment conditions. These concepts are defined in the Law 16/2002 in a similar way as IPPC Directive does.

The application of the Law 16/2002 in Spain presents a special feature due to the fact that the environmental competences were transferred by the Spanish Government to the autonomous regions. So, although this law affects the entire country, the autonomous regions are responsible for its application in the installations located in its own territory. Therefore, the environmental competent authority of the autonomous regions has to handle and grant the environmental permit to a given IPPC installation establishing previously the ELV based on BAT.

In determining BAT, special considerations to items listed in Annex 4 of the IPPC Directive should be paid attention to. Their identification is responsibility of the European Commission (EC), which organises an exchange of information between experts from the Member States of the European Union, representatives from industry and from environmental organisations. This work is coordinated by the European IPPC Bureau (EIP-PCB) and has been mainly divided taking into consideration the productive sectors according to Annex I of the Directive. Each sector is examined by a technical working group (TWG) and it takes around 2 years to complete the work and to produce a so-called BREF (BAT reference document) [5]. The draft BREFs are then examined and discussed in the Information Exchange Forum (IEF), which draws up the final BREFs. The IEF consists of representatives from all Member States, Accession Countries, as well as from Industry and the European Commission.

The epigraph 3.5 of the Annex I of the Law 16/2002 quotes the mineral industries that accomplish the manufacture of ceramic products by firing as follows: "Installations for the manufacture of ceramic products by firing, in particular roofing tiles, bricks, refractory bricks, tiles, stoneware or porcelain, with a production capacity exceeding 75 t/day, and/or with a kiln capacity exceeding 4 m³ and with a setting density per kiln exceeding 300 kg/m³".

The TWG for the ceramic industry sector is organised for the purposes of information exchange under Article 16.2 of the IPPC Directive. On 1st and 2nd December 2003, the EIPPCB, part of the Institute for Prospective Technological Studies of the Joint Research Centre (IPTS) organised the *kick off meeting* in Seville. The TWG agreed that due to the wide variety of ceramic products, the approach to take should focus on summarising the various ceramic sub-sectors with reference to the application of the different types of products in two groups: heavy and fine ceramics [6].

The lack of information at European level has led to different countries to analyze their own state of the art in several

reports [7–11] in order to support and contribute to the process of establishing the identification of BAT for the ceramic sector in the EU. At present, there is a second working draft document on this issue available on line at the EIPPC Bureau website [12]. The final document is expected to be finished in a near future.

BREFs are essential technical tools for complying with IPPC guidelines for both the installations and the administration. Administrative procedure of obtaining (installation) and granting (competent authority) the environmental permit makes necessary a previous analysis of the sectorial existing techniques. Besides a comparison with those techniques that the installation has implemented or other that may be feasible for implementing, from a technical, environmental and economical point of view has to be carried out. In this procedure, it is very important to bear in mind the inherent features of the installation. Accordingly, a method has been developed in Galicia for the largest affected sectors in order to make this task easier for all the involved entities.

The aim of this work is to illustrate this procedure for the application of the IPPC Directive to the heavy ceramic industry in Galicia (NW Spain). Heavy ceramics is a well-established complex sector in this area; however, not so much information on pollution prevention is available in this case. So, this paper revises the special characteristics of this sector in NW Spain and defines the Galician applied method to help heavy ceramic industries and the administration to implement the IPPC philosophy.

2. The ceramic products sector

Generally the term *ceramics* (ceramic products) is used for defining inorganic materials, with possibly some organic content, made up of non-metallic compounds and hardened by a firing process. In addition to clay based materials, today ceramics include a multitude of products with a small fraction of clay or none at all, that can be glazed or unglazed, porous or vitrified [6].

The ceramic industry is included in the European NACE (Classification of Economic Activities in the European Community) Division 26 corresponding to the manufacture of other non-metallic mineral products. This division includes some of the following related groups:

- 26.2 Manufacture of non-refractory ceramic goods other than construction purposes; manufacture of refractory ceramic products.
- 26.21 Manufacture of ornamental ware and household ceramics.
- 26.3 Manufacture of ceramic tiles and flags.
- 26.4 Manufacture of bricks, roof tiles and construction products, in baked clay.

Nevertheless, a division of ceramic products could be carried out according to their key environmental aspects and addressing common issues (such as raw materials, additives, production techniques and product properties) in the following groups [6]:

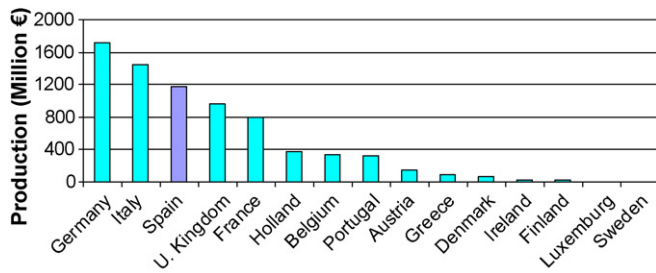


Fig. 1. European ceramic building materials production (million euros) among the EU countries in 2000. Data source [13].

- *Heavy ceramics*, which includes the sub-sectors bricks and roof tiles, vitrified clay pipes, refractory products and calcined clays.
- *Fine ceramics*, which includes the sub-sectors wall/floor tiles, tableware and other household ceramics, sanitary ware and technical ceramics.

The sector of non-metallic mineral products performs the intermediate role of taking minerals that have often been mined or quarried and transforming them into products that can be used in several industries (building industry, civil engineering, metallurgical processes, cement manufacture, glass, incinerators, sanitary ware and tableware). The features of ceramic products include long service life, wear resistance, chemical inertness and low toxicity, fire and heat resistance and, in many cases, aesthetic appeal [11].

This sector has a relatively high reliance on energy, as high temperatures are often required as part of the manufacturing process. Some manufacturers have responded to this challenge by developing and investing in cleaner and more efficient production processes, as well as encouraging the use of recycled materials [13].

Spain is one of the leading countries inside the EU, being third and fourth in the ranking of the manufacture of ceramic building materials (bricks, roof tiles and so on) and refractory products, respectively, as it is shown in Figs. 1 and 2. On a nation-wide scale, the production of building materials in Spain was 19 million tons in 1995. This figure represents the 13% of the ceramic production with an increase of 9 million tons up to 2002, owing to the exceptional dynamism that the construction sector has at present. Although the production of refractory products is lower (339 thousand tons in 1995), it has experienced a similar growth rate for the same period.

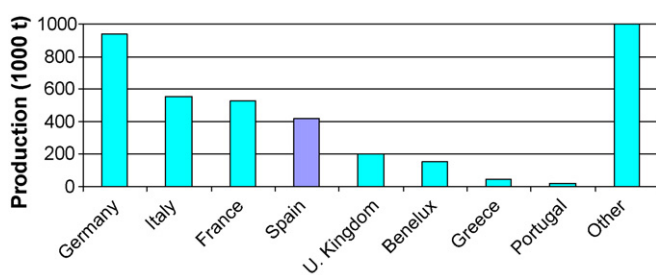


Fig. 2. European refractory products production (1000 t) among the EU countries in 2002 (Benelux and Greece value dates from 1998). Data source [11].

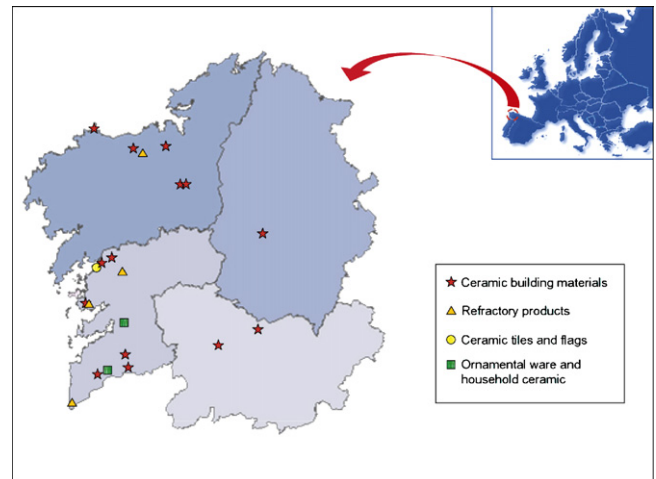


Fig. 3. Geographical distribution of the ceramic sector installations affected by IPPC Directive in Galicia (NW Spain).

Regarding to the Spanish regions, Galicia is the fourth in the national ranking of enterprises with an 8.3% of the total enterprises of the ceramic sector, only overcome by Andalusia, Valencia and Catalonia. Likewise, the number of employed people gets the 7% over the employment in Spain inside the sector and approximately a 4.5% of the net turnover [14]. Nowadays, the sector of ceramic in Galicia is characterised by [15]:

- A heterogeneous group of sub-sectors of small or medium sized enterprises located in Galicia, wherever exist plenty of raw materials of high quality.
- Familiar enterprises at first, although some evolved to anonymous ones and other were absorbed by financial groups of higher economical capacity.
- A good technological level, however most of it is imported.
- Export capacity depending on the quality and the added value of their products.

Nowadays, there are 21 ceramic installations affected by the IPPC in Galicia [16], most of which belong to the group of heavy ceramic industry as shows the map in Fig. 3.

3. Description of the process

Heavy ceramics cover primarily ceramic building materials and refractory products, where clay raw materials are moulded into structural products of both desired shapes. The productive process for the general manufacture of heavy ceramics can be divided into four main stages, as shown in Fig. 4. They include: raw materials preparation, shaping, thermal treatment and post-processing.

In the first stage, the raw materials reception and storage piles take place. Moreover, the crushing, grinding, screening and classifying of them are carried out. The beneficiation and calcination of clays occurs specifically when the refractory products are manufactured. The calcination consists of heating a ceramic material to a temperature below its melting point to liberate undesirable gases or other materials and to bring about structural

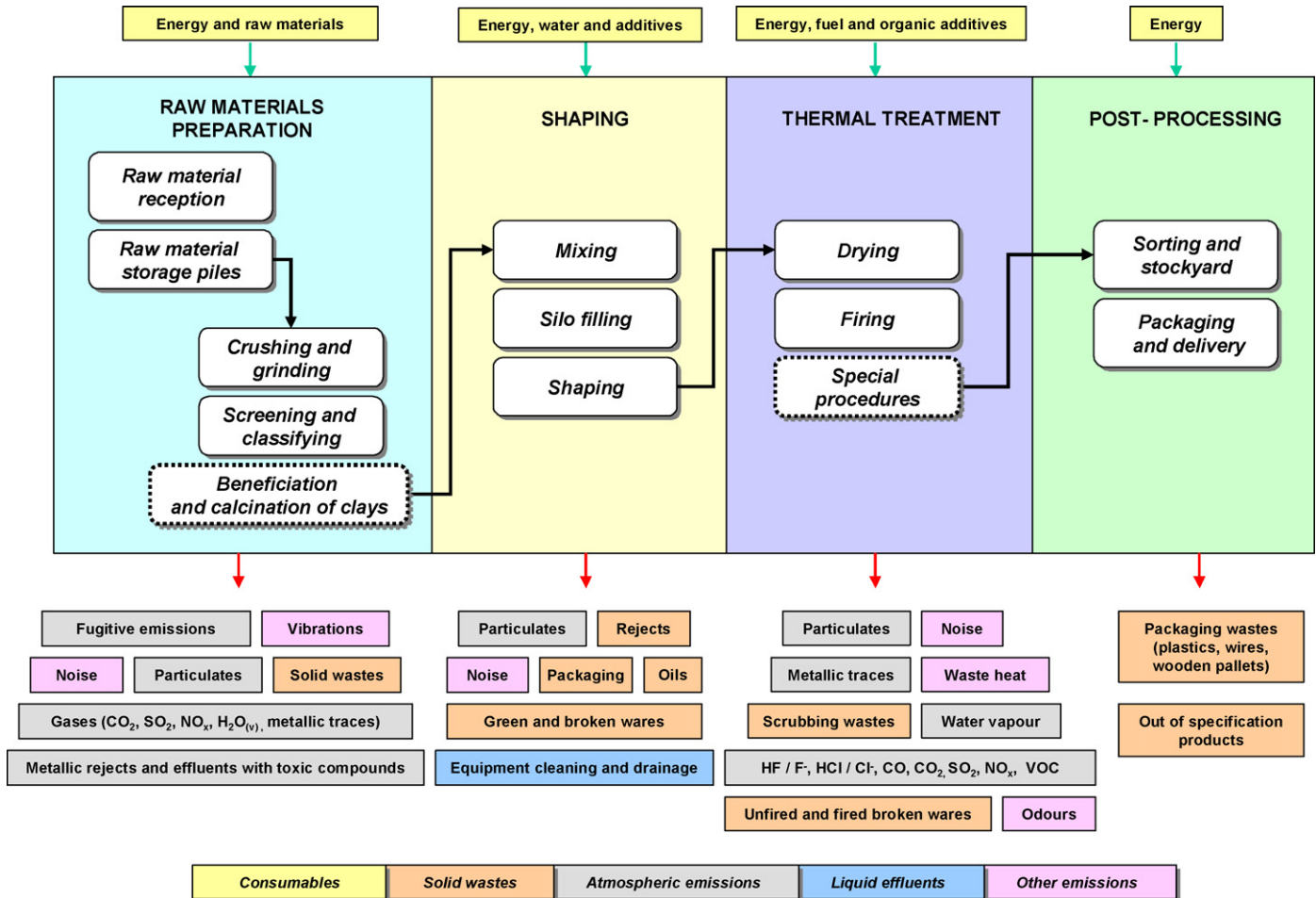


Fig. 4. Flow sheet of the heavy ceramics manufacturing process. It includes inputs (raw materials and auxiliaries) and outputs (environmental impacts). Dotted lines are steps specifically referred to the refractory products production process.

transformation to produce the desired composition and phase product. Calcination usually is carried out in rotary kilns [17].

When the raw materials in the shaping stage have been selected, they are mixed with water to produce a clay body of the required characteristics, auxiliary materials (additives) may be added to the clay body to give certain properties to finished product (shrinkage, porosity, strength, colour and refractoriness) [11]. The clay body is then stored in silos and used for different shaping methods such as soft or stiff-mud moulding, extrusion and pressing, depending on the kind of the material, the water content and the desired product to be manufactured. Shaping operation can be carried out by applying different techniques such as by pressing, extrusion or moulding [18].

Most bricks are made by stiff mud extrusion process, although other by soft mud and dry press processes. The green clay pipes are shaped in vertical de-gasifying extruders, while roof tiles and refractory products are formed by isostatic or hydraulic pressing. Some refractory products are also made by extrusion.

After shaping, the ceramics must be thermally treated. The first step is drying, which can be outdoors or often performed in tunnel kilns. Driers are heated mainly by waste heat recovered from the cooling zone of the kiln, and in some cases by natural gas or fuel oil burners [15], that make the temperature

up to 120 °C over approximately a 24 h period [18]. Secondly, firing takes place in gas heated tunnel kilns predominantly in an oxidizing atmosphere, otherwise in roller, Hoffman or shuttle kilns. The wares to be baked are programmed for different parameters such as firing temperature or time according to a specific product. In this way, bricks and roofing tiles are heated up to a maturing temperature between 900 and 1200 °C, vitrified clay pipes between 1150 and 1250 °C, whereas the wide variety of refractory products range from 1250 to 1800 °C [9]. The manufacture of impregnated refractory shapes using pitch involves special procedures to require a low temperature (250–300 °C) tempering in a kiln [9,11,18].

In the last stage, the finished products enter the storage facility for selection and inspection for quality, packaging, dispatch and distribution [18].

4. Environmental aspects: consumption and emission levels

Diverse raw materials, natural and synthetic, are used to manufacture a broad range of ceramic goods applying various production techniques. Hence, the ample variation in materials, products and their properties, and production techniques, leads

to widely varying levels of consumption and emission [18]. The inputs to the process (raw materials and auxiliaries) and outputs, which represent environmental impacts (consumption of resources, solid wastes, atmospheric emissions, liquid effluents and other emissions), are shown qualitatively in Fig. 4.

4.1. Consumption levels

The Spanish consumption of red clays, which gives the special reddish colour to the brick and roof tile industry, accounts for 33 million tons. This is different from the case of other materials used in the refractory industry, such as alumina, bauxite, magnesite, graphite, silicon carbide, dolomite or refractory clays, depending on the type of refractory product to be manufactured, which jointly represented 519 thousand tons in 2002 [19]. Water is used basically in all ceramic steps, during the preparation of clays slips and clay bodies for shaping, besides in wet beneficiation or grinding processes. The average consumption in manufacturing bricks is about 0.187 m^3 water/t, while for vitrified clay pipes and chromite refractory products is around 150 and 5.0 kg/t , respectively [9]. Referring to cleaning water, liquid effluents during the steps of wet grinding, beneficiation, mixing and shaping are produced. The volume of water used is changeable (about 5 L/t of product, [7]), depending on the applied techniques and the water pressure, even its quantity can be reduced when recycling on the process after settling in tanks is made. Other consumptions come from showers and toilets inside the installation. With regard to energy consumption, both sectors are energy intensive, since a key part of a process involves drying followed by firing to temperatures between 800 and $2000 \text{ }^\circ\text{C}$. The energetic consumption depends on the raw materials used, the manufacturing process and product type in addition to the firing techniques employed. Plant and machinery used for comminution and mixing of raw materials, and shaping of ware, require electrical energy and other equipment, for instance conveyor belts, fan systems or engines, too. Diesel fuel is required for in-works transportation, which may include haulage of raw material from a quarry. In terms of specific energy consumption, the bricks/roof tile sub-sector ranges from 1710 to 2805 kJ/kg , while the energy consumption of the vitrified clay pipes rises as they increase in size [9]. There has been a progressive move to cleaner fuels (natural gas or electricity) and away from coal and heavy oils within the last decades, result of investment into improving energy technology. It brings about a substantial reduction of emissions to air, especially sulphur emission and organic substances. Natural gas is now mostly used and accounts for nearly 90% of the total energy consumption [11]. Other consumptions are represented by the packaging material, with a low percentage over the total from 0.5 to 1.0 g/kg of brick manufactured [9].

4.2. Emission levels

Emissions to air represent the major environmental aspect of the heavy ceramic industry from the point of view of IPPC. Particulate matter (PM_{10}) may arise during handling or processing of raw materials (e.g. grinding operations, screening,

shaping, drying, firing and calcination), specifically in the case of dry materials. Also, fugitive emissions can be produced on the whole installation by broken wares or dusty spillages owing to vacuum cleaning systems are not available on site at all times. Particulates from tunnel kilns may reach values of around 80 mg/m^3 [9]. Gaseous compounds released during drying, calcination and firing are mainly derived from the raw materials (including additives), but the fuels employed also contribute to gaseous pollutants. The compounds concerned are identified in the kiln exhaust gases, which are as follows: sulphur dioxide, oxides of nitrogen, carbon monoxide and dioxide, volatile organic compounds (VOCs), chlorine, fluorine and their compounds and metals as trace elements. The concentration of SO_x (mainly SO_2) in waste gases is closely related to the sulphur content on the raw material and fuels, therefore in brick/roof tile industries 10 – 500 mg/m^3 are emitted depending on the type of fuel employed for firing and in a refractory one is emitted about 10 – 580 mg/m^3 according to silica, magnesia or high alumina bricks manufactured [9]. Nitrogen compounds are present in fuels (mainly solid or liquid types) or in organic additives and they form NO_x during combustion. NO_2 emission is around 20 – 120 mg/m^3 for brick/roof tile industry and gets upper values (30 – 470 mg/m^3) depending on the refractory product type [9]. Carbon monoxide and dioxide not only arise from the combustion of organic matter in the ceramic body, but also fossil fuels and thermal dissociation of carbonates during firing. The CO emission concentration ranges from 10 to 180 mg/m^3 in refractory products [9]. Ceramic raw materials may themselves contain organic matter, and a wide range of organic materials are added, especially in the refractory industry, where pitch impregnation of fired ware is performed to achieve carbon enrichment [11]. During the early heating process, carbonisation of organic compounds occurs with the release of a complex range of VOCs and, later when special treatments as tempering in refractory products take place. Nearly all earth materials contain fractional amounts of fluoride, which substitutes OH^- groups in clays and hydrous mineral, thereby its presence on exhaust gases in 0.5 – 120.0 mg/m^3 [9]. These fluoride emissions can be lower if lime or other fluoride reactive compounds are added to clay mixture due to they enhance the retention of some potential pollutants by forming a more stable compound within the fired ceramic [20]. In this way, most of clays contain trace levels of chloride, originated in marine formation, which gives concentrations over 20 mg/m^3 in emissions [9]. The heavy metal content of most ceramic material is very low, and causes no emission problems [11]. Emissions to water are principally confined to insoluble particulate matter derived from material processing, so can be separated by settling/filtration (often for reuse). Solid wastes are usually minimal, since most wastes generated in ceramic processing are recycled within the process, or find secondary uses (tennis sand, filling material for roadways or quarries, etc., [11]), while other like scrubbing wastes need a specific post-treatment. Other emissions such as noise or odours scarcely have a minimal incidence inside the installations.

Fig. 5 shows an overview of all these potential emissions to different media that bring about environmental effects, which may be harmful to human health or the quality of the environ-

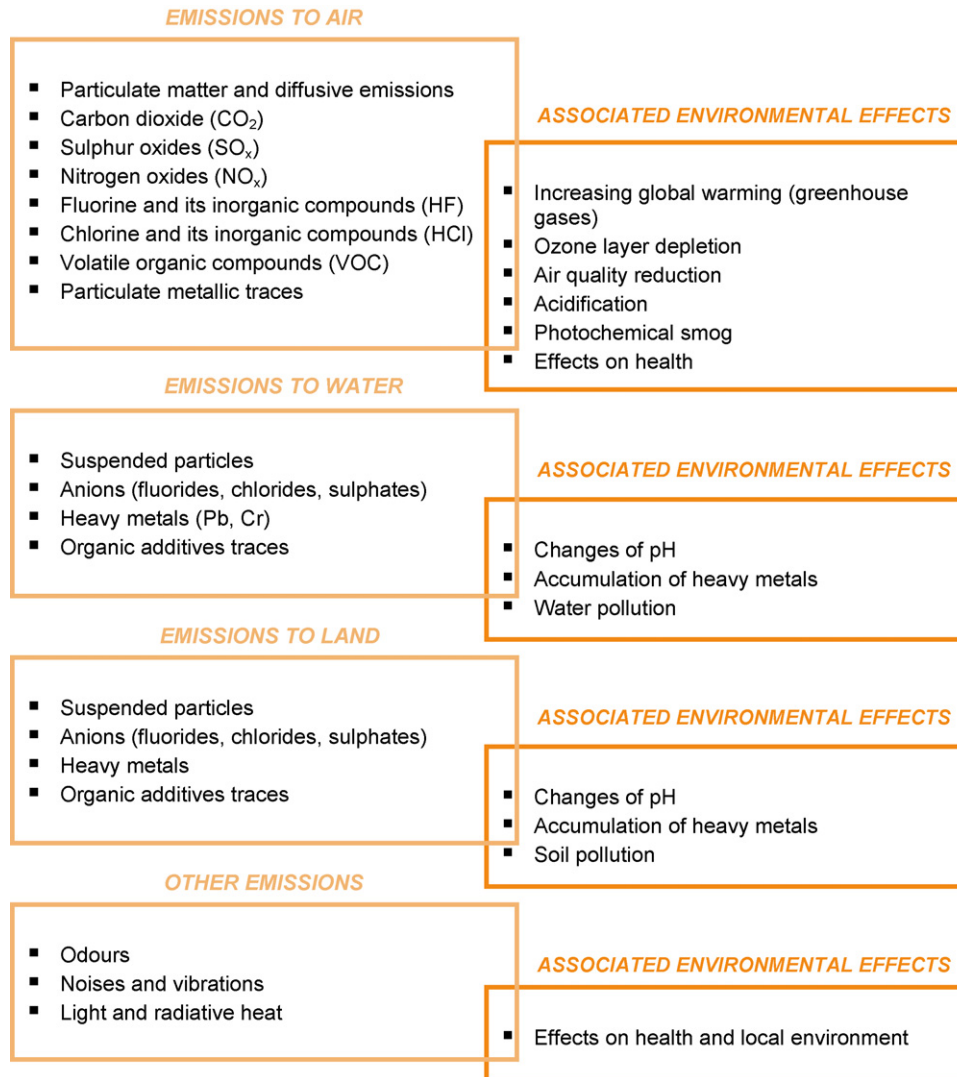


Fig. 5. Summary of potential emissions to different media and their associated environmental effects from heavy ceramic industries.

ment, result in damage to material property, or impair or interfere with amenities and other legitimate uses of the environment.

5. Methodology

Once the industrial process and the aspects related to its potential environmental impacts are clearly defined by stages, it is possible to act upon the specific source of pollution to prevent or reduce the derived polluting effect. To achieve this, a study of all the likely preventive or reduction measures was made, using different sources of background information available on the issue, such as books, journals, reports, personal communications from industry, experts' opinions, etc. It is worth remarking the contribution of several heavy ceramic industries in Galicia. This gives rise to a list of measures for the sector to consider in the initial analysis of BAT.

A definitive inventory of candidate BAT was achieved after undertaking a screening of the previous list taking into account the following aspects for considering in the selection:

- Design or re-design of the equipment used for the thermal treatment of the ceramic pieces in order to minimise the emissions to atmosphere coming from them.
- Low polluting additives and combustibles.
- Wastes minimisation by means of control processes, raw materials inventory control, recycling and reuse, etc.

The measures of the candidate BAT inventory were classified in best environmental practices and techniques (preventive or abatement ones). These latter were treated exhaustively in order to facilitate the establishment as BAT for a given installation, given rise to a technical data sheet for each technique. Then, the information in the data sheet was sorted in the same way that the analysis was carried out, i.e. taking into account the next items along the lines of the methodology followed by the EIPPCB:

- *Environmental aspects*: main environmental impacts to be addressed by the technique.

- *Technical description of the technique.*
- *Benefits or environmental data:* performance data on emissions/wastes and consumption (raw materials, water and energy), apart from emission values for the different pollutants as stated in legislation.
- *Secondary effects:* any indirect effects and disadvantages, as well as details on environmental problems by applying the technique.
- *Implementation:* steps taken for implementation of the technique.
- *Applicability and characterization:* consideration of the factors involved in applying and retrofitting the technique, including useful information on how to operate, maintain and control the technique.
- *Economic aspects:* information on costs (investment and operation) and any possible savings.
- *Plants where it is already implemented:* examples or references plants reported to be used.

For a given installation the analysis of BAT consists of taking into consideration the candidate BAT enumerated in the inventory database and also bearing in mind the age of the installation (if it is existing or new), its technological features, geographical location and environmental conditions, with the purpose of selecting those that are implemented and those which are not. The knowledge of the implemented techniques can be compared with the items of the data sheet, concluding whether it is BAT or not. Regarding the case of not implemented techniques, this methodology represents a feasibility analysis which allows determining whether the chosen techniques can be considered as BAT.

For each specific case, this methodology could be complemented with other existing for the same purpose. Despite they differ from each other, they assist to evaluate from dissimilar points of view the candidate BAT as follows:

- Environmental Impact Assessment (EIA) to evaluate planned projects (including technological process).
- Life Cycle Assessment (LCA), i.e. environmental impact assessment related to the whole life cycle of the product (such as a facility or a technique), including all life steps.
- Sequential procedures such as cross-media guidelines, costing methodology, evaluating alternatives and economic viability of the sector including in the BREF on Economics and Cross-Media Effects [21].
- Combined methodologies, as the carried out by Breedveld et al. [22], where a unique end of pipe technique is analysed by means of a simplified LCA, an eco-efficiency calculation and the additional cost per unit reduction of emissions. Therefore this is a comprehensive procedure to assess whether a technique is considered as BAT or not.
- Methodology based on expert judgement, focusing scores on the technical feasibility, cross media environmental performances and economic feasibility [23,24]. It has the advantage to be simple and convenient, but more profound quantitative analysis is needed to evaluate the impact

of candidate BAT and other methodologies can be also applied.

- Environmental assessment method for cleaner production technologies [25]. It is based on material and energy flows evaluations that use a set of profile indices, including raw material, energy, waste, product and packaging profiles related to the technology under investigation for determining an integrated index.

6. Results

The proposed techniques to be considered as best available techniques or candidate BAT and the environmental aspects they act on, according to the process stages in heavy ceramics installations identified in Fig. 4, are summarised in Table 1. Among them, the *best environmental practices* (BEP) represent an essential part, because by means of their application the environmental impacts can be minimised. Their utility, simplicity, low cost and quick results require first of all changing attitudes on people and processes in the management of any industry.

In general, BEP for a ceramic installation in order to improve the environmental performance should be based as stated below [26,27]:

- Information campaigns to raise public environmental awareness amongst the staff of the installation.
- Records of raw materials, additives, water and energy consumption in the process, as well as the quantity, typology, destination and costs of wastes and their management for setting reduction targets.
- Assess the possible environmental impacts caused by accidents or unexpected emissions.
- Adequately training equipments and materials before operating in order to prevent defective pieces on the start-up.
- Work to a suitable processing speed, due to optimise the production and generate the lowest wastes.
- Optimise heating processes for avoiding waste heat and making best use of combustible.

Moreover, an *Environmental Management System* (EMS) is also a generic technique related to the continuous improvement of environmental performance of an installation that should be determined as BAT.

Technical data sheet are specified in Figs. 6–8, which consider different examples to avoid emissions to land, water and air, respectively. These examples contain practical cases of the procedure as described above and were selected to show different types of techniques:

- Recycling of waste (Fig. 6).
- End-of-pipe improvement by recovering of wastes (Fig. 7).
- Prevention of emissions by process technology optimisation (Fig. 8).

And now the proposed techniques according to process stage are set out.

Table 1
Proposed techniques to be considered as best available techniques and environmental aspects for heavy ceramics manufacture

Process stage	Techniques	Environmental aspect	
Raw materials preparation	<i>Raw materials preparation</i>		
	• Selection of raw materials and additives	Particulates and polluted gases	
	<i>Storage, crushing and grinding of clays</i>		
	• Techniques for reducing fugitive emissions		
	Several techniques	Fugitive emissions	
	• Cleaning systems		
	Centrifugal force separators (e.g. cyclones)		
	Bag filters	Particulates and fugitive emissions	
	Electrostatic precipitators		
	• Techniques for reducing acoustic emissions		
Isolation and acoustic barriers	Noise		
	<i>Beneficiation and calcination of clays</i>		
	• Physicochemical treatment	Effluents and toxic wastes	
	Neutralization and settlement		
Shaping	<i>Mixing and shaping</i>		
	• Water usage optimisation	Reduction of resources consumption (water and energy)	
	• Energetic optimisation		
	• Management of effluents		
	Serial settling tanks	Water process	
	Management of equipment drainage		
	• Management of wastes		
	Reprocessing of green and unfired wares	Green wares	
	Management of empties	Empties	
	Management of used oils	Used oils	
Thermal treatment	<i>Drying and firing</i>		
	• Selection of fuel		
	Utilization of free or low sulphur fuels	Particulates and exhaust gases	
	Utilization of natural gas		
	• Cleaning systems		
	Bag filters with injection of NaHCO ₃	Particulates and acid gases	
	Electrostatic precipitators with injection of NaHCO ₃		
	• Process optimisation		
	Cogeneration system		
	Using calcium-rich additives	Fluorine and its compounds	
	Kiln operation optimisation		
	Exhaust-gas kiln recirculation		
	Heat recovery from flue gases and recirculation to drier	Hot air	
	• Techniques for reducing inorganic compounds		
	Low NO _x burners	NO _x	
	Granulated bed absorber operate with CaCO ₃		
	Cascade-type packed-bed absorber		
Dry scrubber with bag filters or electrostatic precipitator	Acid gases: SO ₂ , HF, HCl		
Honeycomb shaped module absorber system			
Wet scrubber			
• Management of wastes			
Management of scrubbing wastes	Toxic wastes		
	<i>Impregnation and tempering</i>		
	• Techniques for reducing organic compounds		
	Carbonisation gas afterburning inside the kiln	VOC	
	Carbonisation gas afterburning in a counter-travel kiln		
	External thermal afterburning with regenerator columns		
	Catalytic afterburning		
	Adsorption and destruction systems of VOC		
	Special installations	Odours	
	Post-processing	<i>Sorting and packaging</i>	
		• Management of rejects	
Recycling of fired and broken wares		Fired wares	

REPROCESSING OF GREEN WARE	
ENVIRONMENTAL ASPECT	Emissions to land. Rejects and green broken wares produced during the shaping (extrusion or pressing) of the products.
TECHNICAL DESCRIPTION	Reusing of green wastes that are incorporated directly into the product composition during the mixing stage, inside the manufacturing process, with no need of change. In order to prevent contamination, it should be necessary an efficient clay collection system before falling to the floor.
<pre> graph LR A[Mixing] --> B[Shaping] B --> C[Rejects Green broken wares] C --> A </pre>	
BENEFITS / ENVIRONMENTAL DATA	Improvement of natural resources, such as raw materials or energy, with the consequent reduction in waste generation for landfill disposal.
SECONDARY EFFECTS BY APPLYING THE TECHNIQUE	If a good selection of rejects is not made, the paste can be polluted.
IMPLEMENTATION	Implanted.
APPLICATION AND CHARACTERIZATION	The grade of humidity contained in the products (between 18 and 25 %) gives them an enough plasticity and easy shaping for being reincorporated to the process immediately, without any kind of transformation ^(a) .
ECONOMICAL ASPECTS	Cost-effective economically because raw materials are reintegrated to the process. One tile maker is saving about 150.000€/yr as a result of installing a guttering system to collect unfired reject tiles for reuse. The capital cost of the equipment is about 750€ ^(b) .
PLANTS WHERE IT IS ALREADY IMPLEMENTED	Usual practice in Galician installations.

^(a) Source: [7] ^(b) Source: [28].

Fig. 6. Example of technical specifications for the reprocessing of rejects and unbaked ware during the shaping stage of the heavy ceramic manufacturing [28].

6.1. Raw materials preparation

6.1.1. Raw materials extraction/reception

Prior to the extraction of raw materials in clay pits, it is necessary have on mind several measures to minimise the impacts they produce on the environment and how optimise their usage, as follows [18]:

- Adequately selection of raw materials taking account of the emission limit values and wastes they cause.
- Maintain an inventory covering the principal types of raw materials and additives used.
- Annually review alternatives for the principal types of raw materials and additives used with regard to their environmental impact.
- Substitution of organic additives for inorganic ones.
- Control the specification of raw materials and additives used in order to minimise any potential environment impact.
- Reduce the usage of chemical reactives and other polluting materials.

6.1.2. Storage, crushing and grinding of clays

The general measures to be employed during the storage, crushing and grinding of clays related to avoid fugitive emissions are the following [18]:

- Ensure that, where there is vehicular movement, storage areas have a consolidated surface which is kept in good conditions.
- Wet stockpiles where necessary to minimise dust emission and install fixed water sprays for long term stocking areas if appropriate.
- Provide adequate protection against wind whipping.
- Clean all process buildings regularly, according to a written maintenance programme to minimise fugitive emissions.
- Use closed and independent warehouses to control emission sources.

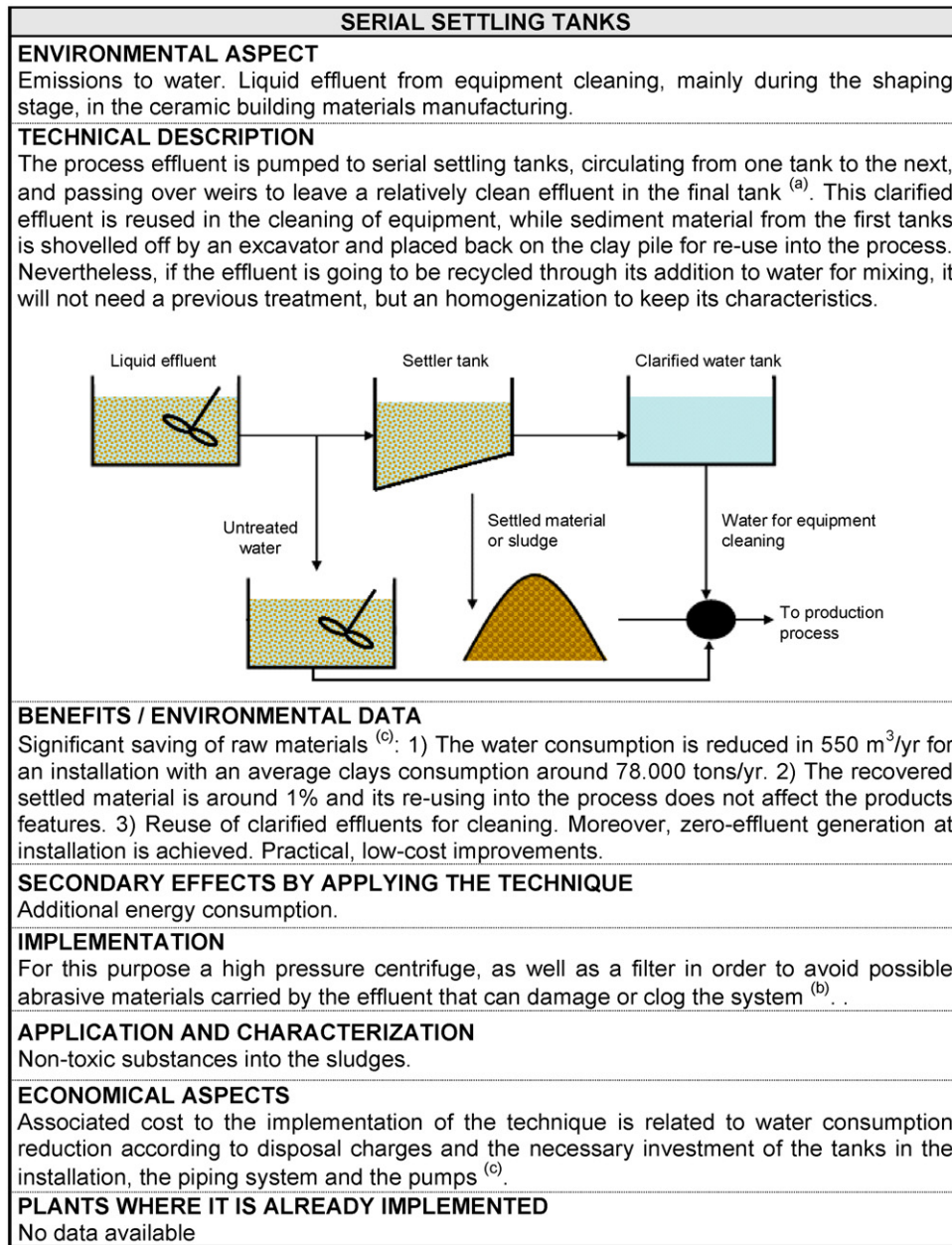
The techniques for the efficient removal of particulate matter and dust emissions are:

- Cleaning systems of particulates: centrifugal force separators, bag filters and electrostatic precipitators [7,9,18,27].
- Isolation and acoustic barriers for the minimisation of acoustic emissions.

6.1.3. Beneficiation and calcination of clays

The measures to be considered during the beneficiation and calcination of clays are the following:

- Optimisation the calcination process: residence time, length of the kiln, time–temperature profile, etc.



^(a) Source: [7] ^(b) Source: [11] ^(c) Source: [29].

Fig. 7. Example of technical specifications for the recovering and recycling of waste effluents coming from heavy ceramic installations with serial settlement [29].

- Installation of cleaning systems of particulates and acidic gases coming from the calcination kiln: centrifugal force separators, bag filters and electrostatic precipitators, which include alkaline reactives to neutralise the acidic emissions.
- Segregate solid and toxic wastes rejected and stored adequately in special containers clearly labelled in secured places.
- Segregate toxic effluents when the purification of raw materials is carried out by wet processes. These liquid effluents need a physicochemical treatment with the stages of homogenisation, pH adjustment, coagulation, flocculation, sedimentation, treatment and sludge stabilization. Later, a sludge analysis is needed and they are sent to disposal landfill if they have not

hazardous heavy metals to force them to a safety storage tank [8].

6.2. Shaping

6.2.1. Mixing and shaping

The techniques to be considered for reducing the emissions are the following:

- Water usage and energetic optimisation.
- Mechanical dosage systems for the mixture of raw materials and water, improving resources and quality at maximum.
- Installation of isolation and acoustic barriers for minimising and/or preventing acoustic emissions and vibrations from machinery.

EXHAUST GAS KILN RECIRCULATION	
ENVIRONMENTAL ASPECT	
Fluorine and its compounds emissions to air.	
TECHNICAL DESCRIPTION	
<p>Most clays contain fluorides of varying concentrations that are released during firing above 600 °C and emitted into the atmosphere mainly as hydrogen fluoride (HF). The technique consists on the installation of a recirculation system in the preheat section of a tunnel kiln where the temperature ranges between 150–400 °C. The system recirculates gases from one zone to an adjacent, higher-temperature zone. This increases the turbulence within zones and the residence time of the flue gases where the fluoride absorption takes place. The absorption/emission process is as follows ^(a):</p> <ul style="list-style-type: none"> • The clay product enters the kiln. During the firing cycle, fluoride in the clay body reacts with moisture in the kiln atmosphere and forms hydrogen fluoride. A small proportion of this reacts with airborne particles to produce other fluorides. • The gaseous and other airborne fluorides are mixed with, and transported by, the counterflowing kiln gases. They flow back towards the stack near the kiln entrance. • The product is preheated by its interaction with the kiln gases and absorbs a proportion of the airborne hydrogen fluoride as calcium fluoride in the clay body product. • The product leaves the kiln now contains fluoride from two sources, that inherent in its composition and that absorbed from the kiln atmosphere within the product. 	
<p>The diagram illustrates the fluoride recirculation process in a tunnel kiln. It is divided into a 'Preheat zone' on the left and a 'Firing zone' on the right. A 'Push' arrow indicates the direction of material flow from left to right. In the firing zone, 'Fluoride inherent to the clay' (red arrows) and 'Fluoride retained in the brick' (red arrows) are shown. 'HF emissions' (orange arrows) are shown moving from the firing zone back towards the preheat zone. In the preheat zone, 'HF absorption' (orange arrows) is shown where fluoride is taken up by the clay. 'Fluoride in stack gases' (orange arrows) is shown exiting the preheat zone. A legend indicates that red arrows represent 'Fluoride within the brick' and orange arrows represent 'Fluoride in the kiln atmosphere'.</p>	
BENEFITS / ENVIRONMENTAL DATA	
<p>Around 65% reduction in fluoride emission (from 25 mg/m³ to below 10 mg/m³), without increasing other pollutants emissions, by recirculating a third of the exhaust gases. The recirculation system has also improved the top-to-bottom temperature distribution of the kiln. Compared to an end-of-pipe technology ^(b): 1) Reduce the demand for limestone (used as a reagent in waste gas scrubbers) and consequently reduce the environmental impact of its quarrying and the associated transport operations. 2) Generate less waste for disposal to landfill. 3) Eliminate additional process energy requirements with their associated release of carbon dioxide, one of the major greenhouse gases.</p>	
SECONDARY EFFECTS BY APPLYING THE TECHNIQUE	
<p>An excessive limestone addition can cause the so-called brick efflorescence ^(c). Also, due to added limestone decarbonisation during firing in the kiln, carbon dioxide emissions could be incremented.</p>	
IMPLEMENTATION	
<p>The implementation and the running costs of process modification are installation specific and, they will depend on the design and kiln operation.</p>	
APPLICATION AND CHARACTERIZATION	
<p>No more than 5% limestone should be incorporated to the clay mixture during manufacturing process to avoid brick efflorescence ^(c). Fine limestone or chalk (CaCO₃) is a fluoride reactive compound that can react and therefore reduce fluoride emissions. Its particle size distribution is crucial for a successful fluoride capture.</p>	
ECONOMICAL ASPECTS	
<p>Low capital cost (45.000 €) and running cost (2.250 €/yr). No additional maintenance costs. Saving is around 450.000 € with regard to an end-of-pipe technology.</p>	
PLANTS WHERE IT IS ALREADY IMPLEMENTED	
<p>Ibstock Building Products Ltd. (United Kingdom)</p>	
<p>^(a) Source: [20] ^(b) Source: [29] ^(c) Source: [11].</p>	

Fig. 8. Example of technical specifications for the reduction of fluoride air emissions through process optimisation by exhaust gas kiln recirculation.

- Cleaning systems of particulates: centrifugal force separators, bag filters and electrostatic precipitators.
- Reuse of green and unfired wares within the process, saving raw materials.
- Management of packaging and used oils.
- Recycling of industrial effluents (equipment drainage and cleaning), treating them first if necessary and when this is not practicable, they should be recycled to another part of the pro-

cess which has a lower water quality requirement. Moreover, sludge can be reincorporated to the mixture of raw materials.

6.3. Thermal treatment

6.3.1. Drying and firing

The techniques to be used for optimising the process and reducing the emissions are [9,11]:

- Selection of the fuel used, with low levels of pollutants, for example free sulphur fuels or natural gas.
- Installation of particulates and acidic gases cleaning systems coming from the kiln: bag filters or electrostatic precipitators, which injection of alkaline reactives to neutralise and minimise the acidic gases.
- General process optimisation of (i) the kiln parameters: altering the time–temperature profile, reducing the air flow through the kiln, increasing the turbulence in the preheat zone, increasing the interaction between the product and the flue gas and utilising the flue gas, each parameter gives a different percentage of reduction in fluoride emissions, as Envirowise states [20,30]; (ii) fluoride reactive compounds; (iii) cogeneration systems; (iv) flue gases internal recirculation and recovery of waste heat to drier.
- End-of-pipe technologies for reducing inorganic compounds existing when the abatement measures at source are not enough in order to achieve the emission values by law: scrubbers, low NO_x burners, etc.
- Management of scrubbing wastes.

6.3.2. Impregnation and tempering

When refractory products are required to work in extremely hostile working environments it is necessary to carry out special treatments by means of impregnating fired ware with petroleum-based pitch. In that case, several measures for minimising organic compounds to atmosphere are shown as follows:

- Special installations where the refractory products impregnation is made for preventing odours emissions.
- End-of-pipe technologies for reducing volatile organic compounds released from pith during the firing [18].
- General optimisation of the tempering kiln parameters.

6.4. Post-processing

6.4.1. Sorting and packaging

The measures adopted in the last stage are referred to the management of rejects (fired and broken wares) and/or products out of specification, whose recycling as secondary raw material is the best option.

6.5. Integrated technologies and techniques

These are based on systems that utilize the natural resources very efficiently, with entry flows of low environmental costs, generate little or no residues, recycle them when they exist and release non-toxic effluents. Moreover, the industrial symbiosis

and industrial ecosystems must be also taken into account, this is, the integration of the plant in industrial complexes where by-products of matter and energy are employed as secondary raw materials and not as wastes.

The integrated technologies and techniques that might be carried out in a ceramic installation are:

- Improvement of vegetal oil wastes coming from food industry for the combustion process in the ceramic products thermal treatment.
- Improvement of contaminated soils, particularly those of argillaceous nature, with an identical composition to the raw materials used in the ceramic industry that manufactures structural ceramics [31].
- Ceramic valorisation of wastewater treatment plant sludge [32], wastes from heavy fuel-oil spillages on the sea, red mud from the alumina manufacturing [33], etc.

7. Concluding remarks

In this work the application of the IPPC Directive to the heavy ceramic industry in Galicia (NW Spain) is illustrated. Firstly, a structured overview of the situation and the special features of the sector in this region are shown. The productive process for ceramic building materials and refractory products and their related environmental aspects are outlined. This latter feature was analysed qualitatively in order to provide with information to carry out the identification of candidate BAT. At the time of determining specific BAT for these installations, considerations according to Annex IV of the IPPC Directive have been taken into account. Furthermore, the local environmental conditions, geographical location and distinctive technical characteristics of each installation will be regarded. In this sense, a general standard structure of technical data sheet or specifications was used for describing each technique in order to help installations to assess candidate BAT by themselves. A list of 37 recommended BAT was described and examples were selected to show the simple and practical procedure described in our work and enlighten different types of prevention and mitigation techniques in the heavy ceramic industry (recycling of waste, end-of-pipe improvement and prevention of emissions by process optimisation).

As far as the proposed techniques implemented in the Galician sector are concerned, few of them were implemented in heavy ceramic existing installations and there is no evidence to those regarding the management of water emissions because these effluents are not produced.

The performed study is part of a designed support guide for the ceramic sector that includes an application form to submit the environmental permit [34]. Its goal is to help applicants (owners of the installations affected by IPPC) to identify the necessary process related information, being also useful both for the technical equipments in charge of gathering data, regulators in the judgement of that documentation and also public in general. Then, this allows the integration of all required information in the administrative procedure to be available at the time to solve the permit, so the permits can be granted faster and more easily.

In this way, all of them are taking into account the study that this paper reports in accordance to the requirements of the integrated pollution and prevention control policies.

Galicia is relatively short of requests for the environmental permit with respect to the nearly 300 IPPC installations identified in this area. Nowadays, there are barely 46 installations between new and existing ones whose administrative procedure is still in progress, even though 10 have just granted the permit. Only one of this latter is a ceramic roof tile manufacturing industry and other two heavy ceramics that have submitted the application, however most of the others are preparing the documentation.

A future work complementary to this envisages the possibility of an energy and material flow analysis to detect 'hot spots' of the main environmental impacts and the most polluted stages in the ceramic industry. In this sense, a case study applied to a roof tile ceramic manufacturing plant in Galicia has just been finalized [35]. This valuable and significant information could allow not also cleaner modifications in the operational ways of the processes, but also the selection of both preventive and best environmental practices, as well as control techniques to minimise the polluted effects derived from all kind of installations.

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