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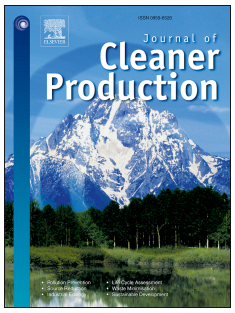

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PII: S0959-6526(19)31815-3

DOI: https://doi.org/10.1016/j.jclepro.2019.05.279

Reference: JCLP 17044

To appear in: *Journal of Cleaner Production*

Received Date: 31 January 2019

Revised Date: 22 May 2019

Accepted Date: 24 May 2019

Please cite this article as: M M, S C, J L, A Ferná, E Muñ, E S, R A, A I, Environmental performance of alternatives to treat fly ash from a waste to energy plant, *Journal of Cleaner Production* (2019), doi: https://doi.org/10.1016/j.jclepro.2019.05.279.

Environmental performance of alternatives to treat Fly Ash from a Waste to Energy plant

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Keywords: carbonation; fly ash; incineration; life cycle assessment; municipal solid waste

ABSTRACT

Incineration has been adopted by many developed countries as an alternative to treat municipal solid waste due to its capacity to reduce the amount of waste and recover energy. Waste to energy plants produce two waste streams: bottom ashes and fly ashes (FA). FA are classified as hazardous waste, and they cannot be utilised or landfilled without prior treatment. Stabilisation with cement solidification is the most used method to treat FA because it achieves the immobilisation of pollutants at a relatively low cost. However, the accelerated carbonation of FA, which allows the encapsulation of certain mobile metals under alkaline conditions, has recently been proposed as an alternative to the solidification/stabilisation process. To determine the environmental performance of FA stabilisation and carbonation, a life cycle assessment (LCA) was conducted. The LCA results of the carbonation and stabilisation processes were compared, and multiple carbonation scenarios were analysed: carbonation with different CO_2 sources (incineration flue gas and flue gas from the combustion of natural gas), and different pressures (1 to 5 bar) and percentages of CO_2 excess (10%, 55 % and 100 %) in the flue gas stream.

Stabilisation had higher environmental impacts than carbonation due mainly to cement production and consumption. The best operating conditions of the carbonation process were found at flue gas pressures between 3 and 5 bar, since the total energy consumption decreases as the pressure increases. Moreover, the environmental benefits associated with the substitution of electricity from the grid mix made the scenarios based on the combustion of natural gas perform better than those that use the incineration gases as a CO_2 source.

1. Introduction

The unstoppable increase of municipal solid waste (MSW) generation in the last decades reflects the consequences of the production and consumption patterns and the population growth, which is increasing tremendously at a rate of 2.035% yearly (Khandelwal et al., 2019). In fact, the global MSW generation levels in 2016 reached 2.01 Gt, and in 2050 this number could grow to 3.40 Gt. This represents a generation rate of 0.74 kg per person and day. However, this global generation rate ranges widely, from 0.11 to 4.54 kilograms kg per person and day (Kaza et al., 2018). The improper management of MSW contributes to environmental problems since the sector represents between 3% and 4% of total global greenhouse emissions. In addition, the CH_4 from landfills and wastewater collectively accounted for about 90% of waste sector emissions (Babel and Vilaysouk, 2015). Therefore, both developed and developing countries face the challenge of sustainably managing MSW (Halkos and Petrou, 2018). In Europe, although the European Directive 2018/851 (EC, 2018) is promoting the reduction of waste generation and disposal and the connection of resource use and waste residuals (Tsai, 2016), a great amount of MSW still ends ups in landfills (Margallo et al., 2018a). However, as Figure 1 displays, the total amount of MSW landfilled in Europe in the period 1995-2016 has diminished 59%, whereas waste incineration has grown steadily 50% in the same period (EUROSTAT,

2018). These data can be associated with the growth of legal restrictions, such as the Directive on Landfills (EC, 1999), which aims to reduce the amount of MSW landfilled, and the Waste Framework Directive (EC, 2008), currently amended by Directive 2018/851 (EC, 2018), which set several targets for recovery, recycling and collecting MSW to minimise environmental impacts.

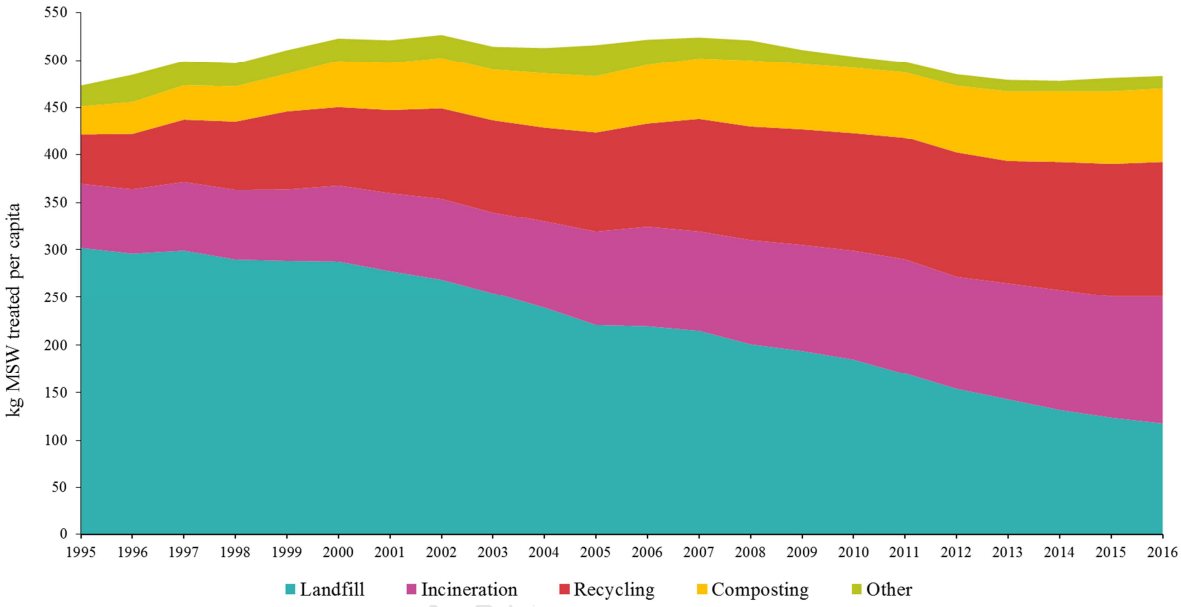


Figure 1. Amount of waste generated at EU-28 level and the amount of waste by treatment category (EUROSTAT, 2018).

Waste-to-Energy (WtE) technologies (e.g. incineration, gasification, pyrolysis, anaerobic digestion) are technologies capable of harvesting the energy content of waste and transforming it into a secondary energy source. incineration, which is capable of reducing the volume of waste by up to 90%, or 75% in weight (Li et al., 2019), is the most widespread WtE technology. These types of WtE plants produce two waste streams: bottom ashes (BA) and fly ashes (FA).

BA constitute the largest fraction (around 80-90%) of the residues generated in the

incineration process (Li et al., 2012; Wongsa et al., 2017). Between 10 and 35% of the combusted MSW becomes BA (Vehlow and Seifert, 2012; Kuo et al., 2007), whereas FA represent between 3 and 5% of the mass of MSW that is incinerated (Joseph et al., 2018; Nam et al.; 2012). Although FA are produced in a lower amount than BA, they are classified as hazardous waste, since these residues collected from the flue gas purification system contain heavy metals, dioxins and other pollutants (Sun et al., 2016). In a recent study (BIO by Deloitte, 2015), the impact of different classification approaches for hazard property "HP14" was assessed for incineration FA with a European Waste List code 19 01 13* (FA containing hazardous substances) and 19 01 14 (FA other than those mentioned in 19 01 13) (Quina et al., 2018). The main problem of FA is that due to their hazardous characteristics, they cannot be utilised of even landfilled without prior treatment. Therefore, the application of a certain treatment may have two approaches: ensuring that the FA can be landfilled in non-hazardous landfills or improving the possibilities of their valorisation (Margallo et al., 2018b) with the aim of fostering the circular economy concept (Margallo et al., 2015). At present, separation processes, solidification/stabilisation (S/S) and thermal methods are the three main techniques applied to treat FA. Among S/S alternatives, stabilisation with cement solidification is still the most widely used, due to its inexpensive and easy application. During the cement solidification process, FA and cement are mixed with water in an appropriate proportion, and the hazardous content of FA is immobilised after the cement hydration reaction (Sun et al., 2016). However, the long-term stability of the cement-solidified FA is not a negligible issue due to the high content (10-20%) of soluble salts (Li et al., 2018). Moreover, the cement sector is an energy intensive industry that accounts for roughly 5-8% of global CO_2 emissions (Mikulcic et al., 2016). Recent studies have investigated new FA treatments, such as carbonation. The low solubility of the

carbonate compounds produced in the carbonation reaction facilitates the reuse or deposition of the carbonated FA in non-special landfills (Grandia et al., 2011). Furthermore, the carbonated FA could act as a CO_2 sink. (de Boom et al., 2014). Therefore, this technology could be a good alternative to handle the technical and environmental drawbacks of other FA treatments. However, the analysis of the environmental performance of this treatment is mandatory to have a good description of the advantages and disadvantages of the system. One of the most widespread techniques for this type of analyses is the Life Cycle Assessment (LCA) methodology. This tool allows assessing the inputs, outputs and the potential environmental impacts and resources used throughout a product´s life cycle, and it reveals cross-media issues (Margallo et al., 2013; Laso et al., 2016).

In recent years, several studies have focussed on the application of LCA to analyse the treatment of residues from MSW incineration. Boesch et al. (2014) developed an LCA model of MSW incineration and new technologies for metal recovery from ash residues. Other authors have assessed the environmental impact of the management of FA by means of cement stabilisation (Yin et al. 2018). Alternatives to the disposal of the stabilised FA in landfill, such as the reuse of FA as bricks and as an alkali in the Waelz process of steelmaking (Huang and Chuieh, 2015), and the acidic FA extraction with integrated zinc recovery (Huber et al. 2018) have also been analysed. Furthermore, Huber and Fellner (2018) and Huang et al. (2018) integrated LCA and an economic assessment to evaluate the valorisation of FA.

On the other hand, the feasibility of the carbonation technology as a FA treatment has not been studied to such a great extent, and the treatment of FA with the carbonation technology at large scale has not been reportedSeveral studies have investigated the optimal

operating conditions for the accelerated carbonation of MSW incineration FA (Li et al., 2007). Baciocchi et al. (2009) compared different reaction routes for the carbonation of air pollution control residues of an MSW incineration plant, and Jianguo et al. (2009) studied the influence of several operating parameters of the carbonation process of the FA from MSW incineration on the stabilisation of heavy metals. The leaching properties of MSW incineration FA were also studied by Boom et al. (2014).

The environmental assessments of carbonation processes found in the literature are primarily aimed at the treatment of steelmaking slag; Chang et al. assessed the carbon footprint of the carbonation of different types of steelmaking slag, whereas Xiao et al. (2014) performed a more comprehensive LCA of alternative carbonation routes. To the best of the authors knowledge, the environmental assessment of the carbonation of FA derived from the incineration of MSW has only been addressed by Margallo et al. (2018b).

Hence, the technical and environmental aspects of FA carbonation are analysed in this paper. The study includes the experimental set up of a carbonation process that provides a representative life cycle inventory. The experimental results of carbonation were compared with previous bibliographic data of FA stabilisation (Margallo et al., 2014a), and the LCA of both FA treatments was performed in order to propose measures to improve the FA management sector.

2. METHODOLOGY

This paper evaluates the environmental performance of several methods to treat FA from a WtE plant. The work applies the LCA methodology proposed in the ISO 14040 (ISO, 2006) combining the use of bibliographic and experimental data. This methodology is based on a four-phase process: i) definition of goal and scope; ii) life cycle inventory; iii) life cycle impact assessment and iv) interpretation (Margallo et al., 2014b).

2.1.Goal and scope

Goal and scope definition is one of the most important phases of the LCA methodology, because the choices made at this stage influence the entire study (De Marco et al., 2017). The goal of this study is to evaluate the treatment of FA from a WtE plant located in the Cantabria Region (North of Spain). Spain has 10 incineration plants, four incinerators located in the Northeast (Catalonia), three in the North and Northwest (Cantabria, Galicia and Basque Country), one in Madrid, one in the Balearic Islands, and one in Melilla. Most WtE plants are located in the North of Spain due to the limited availability of land in this area, and the amount of landfill leachate that is generated because of the intensive rainfall. In particular, in Cantabria an average of 113,338 t of MSW are incinerated per year, generating around 4,500 t of ashes (Margallo et al., 2012).

The analysis was structured in three blocks: i) the analysis of FA carbonation at different pressures and CO_2 excess in flue gas, ii) the assessment of FA carbonation using different CO_2 sources and iii) the comparison of FA carbonation and stabilisation. To determine the dimensions of the carbonation reactor (on which the energy consumption of the process depends), the following scenarios were studied: 10 %, 55 % and 100 % excess of CO_2 in the flue gas at the outlet of the reactor, and flue gas pressures of 1 to 5 bar. The comparison of FA carbonation and stabilisation has to be made based on the same reference unit. The combustion process of natural gas is considered a part of the life cycle of the FA carbonation and thus, the environmental impacts related to the generated electricity are considered a secondary system function.

2.1.1 Function and functional unit

The system function is to treat the hazardous waste generated in a WtE plant. Therefore, the functional unit (FU) should reflect this function. According to Lundie and Peters (2005),

the FU is the definition of the functional outputs of the product system, which provides a reference to which the inputs and outputs can be related. Most LCA studies have been related to the manufacture of products and thus the FU usually makes reference to the amount of product. However, in the study of waste management systems, the function is not to produce anything, but to provide the service of handling waste in a given area (White et al., 1997). In this type of studies the most common FU is the amount of waste produced in a specified year and geographical area. In this case, the amount of FA produced in 2014 in Cantabria, 4,655 t (GOBCANT, 2016), was selected as the FU in order to compare the environmental performance of the solidification and carbonation treatments.

2.1.2 System description and boundaries

Figure 2 depicts the three scenarios based on carbonation (scenario a.1 and a.2) and stabilisation (scenario b). The analysis included the incineration process and the treatment and end-of-life of the FA. The WtE plant is a common subsystem in all the scenarios, and thus, it could be neglected in a comparative analysis. However, due to the use of the flue gas generated in the combustion process as a CO_2 source in scenario a.2, the impacts associated with the WtE plant were analysed in the three scenarios. Based on a previous study of the authors (Margallo et al., 2014a), the combustion and flue gas treatment were considered as a single subsystem. The consumption of fuels (natural gas or diesel), ancillary materials, and reagents, as well as the amount of MSW treated, are the main inputs of the process, whereas the outputs include the energy produced and the waste and air emissions generated. BA are subjected to a magnetic separation that recovers c.a. 10% of material as scrap, and the remaining inert slag is sent to a landfill. The production of steel with scrap is outside of the system boundaries. Regarding the FA treatment, a detailed description of both scenarios is provided below:

Scenario a: FA carbonation. Accelerated carbonation improves the chemical properties of alkaline waste and allows it to be used for new industrial purposes or disposed of in non-special landfills. With this process, heavy metals cannot be removed, but they can be separated and/or stabilised within other matrices (Grandia et al., 2011). Carbonation involves the dissolution of CO_2 in water at initially alkaline conditions. This causes a decrease in the pH of the leachate, and calcite to precipitate until the material is in equilibrium with CO_2, because of the change in the solubility of the metals. The carbonation mechanism is often described as at least a two-step process including a prior CO_2 absorption in water, followed by the carbonation reaction in aqueous medium (Rendek et al., 2006). The life cycle inventory of the carbonation process was derived from the experiments performed at laboratory scale. The experimental setup consisted of a 2 L jacketed glass reactor with an agitation speed of 45 rpm and operated at 20^0C and 1 atm. Pure CO_2 was bubbled through the upper part of the vessel, which contained 100 g of FA from a Spanish WtE plant with a 44.34% CaO content. The carbonation experiments were carried out for different liquid to solid ratios (L/S) varying between 3 and 8 $L \cdot kg^{-1}$. The pH was continuously monitored; it was initially above 12, and it dropped below 7. The stabilisation of the pH, which took approximately 90 minutes, indicated that the reaction was over. The carbonated FA were subsequently subjected to vacuum filtration and dried in an oven at 105 ^{0}C. Leaching tests were performed on the sample in accordance with European Standard EN 12457-4 (EN, 2002). The chloride content in the leachate was quantified following ISO 9297 (ISO, 1989), based on Mohr's method, whereas the concentration of lead and zinc in the leachate was measured by means of atomic absorption spectrophotometry. On the other hand, a thermogravimetric analysis determined the amount of CO_2 retained in the carbonated FA. The results, compiled in Appendix A, show that the

L/S ratio that led to the minimization of the chloride, lead and zinc content in the leachate is 6 $L \cdot kg^{-1}$. Higher L/S ratios might hamper the diffusion of CO_2 in the liquid, and lower ratios may not provide enough water for the required amount of CO_2 to dissolve, given its low solubility. Thus, the system was modeled for this L/S ratio. This scenario includes FA carbonation, a filter press to reduce the amount of water in the ashes, the treatment of the effluent and the disposal of the carbonated ashes. The generation and consumption of CO_2 and water are the main inputs of the carbonation process. Based on the CO_2 source we can distinguish between scenario a.1 and a.2. In the former, CO_2 is obtained from the combustion of natural gas, which generates electricity. In scenario a.2, CO_2 from the flue gases of MSW combustion is used as raw material, reducing consumption of natural gas with respect to scenario a.1.

Scenario b: FA stabilisation. In this process, the hazardous components of MSW are immobilised in a solid matrix. That is, they are physically adsorbed or encapsulated, or a change in the physicochemical form of the pollutant components occurs (Margallo et al., 2015). The FA stabilisation scenario includes the extraction and transportation of raw materials to the solidification process. It employs a mixture of water (23.08 %), cement (17.75 %), and ashes (59.17 %). The stabilised mixture is sent to a landfill.

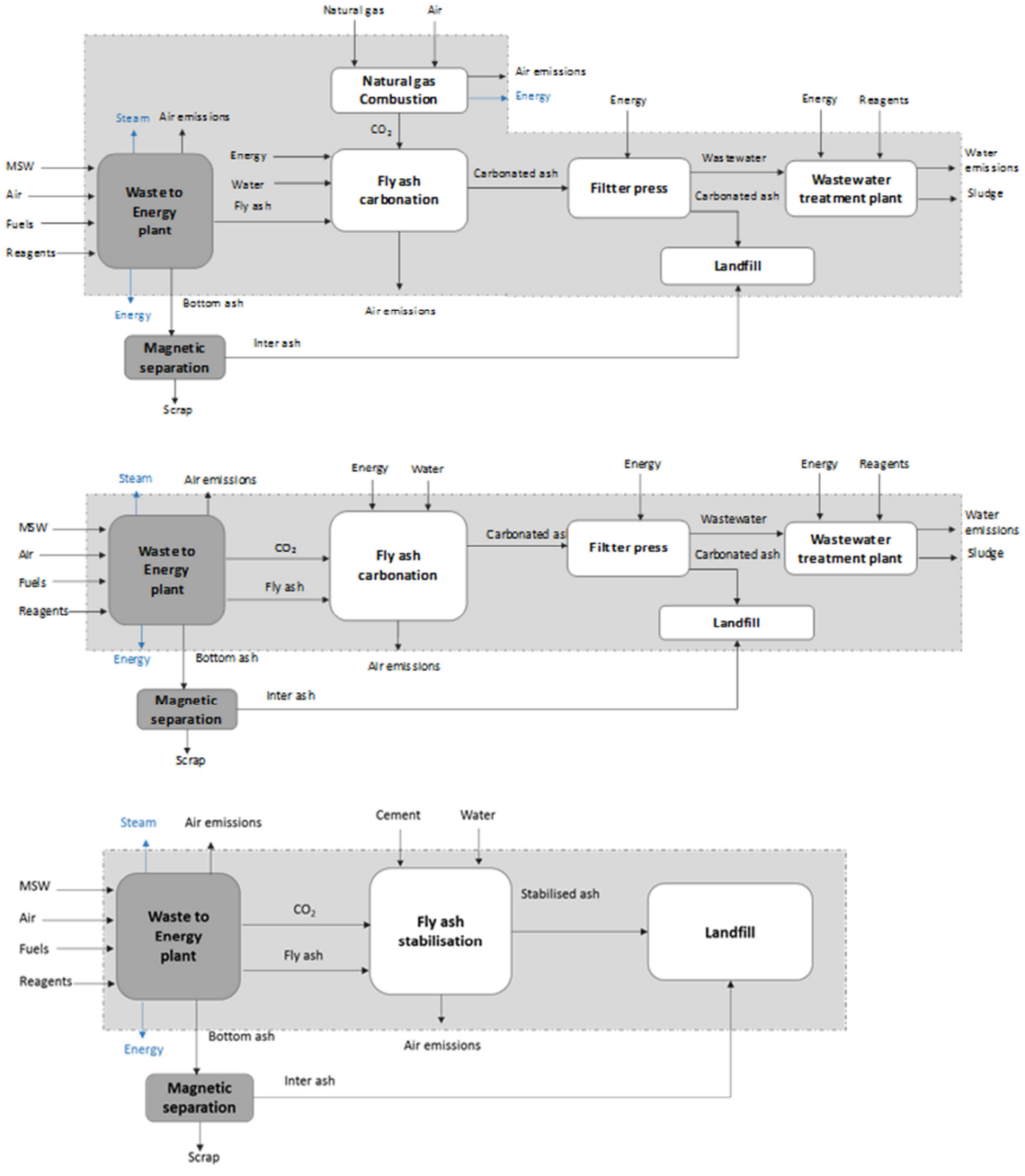


Figure 2. System boundaries of scenario a.1: FA carbonation based on natural gas combustion, scenario a.2: FA carbonation based on the use of WtE flue gases, and scenario b: FA stabilisation.

2.1.3 Allocations

Most industrial systems are multifunctional systems, that is, they comprise processes in which several products and co-products or services are generated. According to the LCA methodology, allocating the environmental burdens of a system between its multiple functions should be avoided by i) dividing the unit process into two or more sub-processes (which cannot be applied to this case study) or ii) expanding the system to include the additional functions related to the co-products (ISO, 2006).

This last method, which was applied in this study, is based on the subtraction of the environmental impacts of the secondary system function from the overall environmental impacts of the system (Thomassen et al., 2008).

On the one hand, MSW incineration involves waste treatment and energy and steam production, providing the system with two additional functions. This problem was handled through system expansion by subtracting the function of the alternative system (energy and steam production) from the system under study. In this study, the Spanish electric power mix and the production of steam from natural gas included in the thinkstep databases (thinkstep, 2017) were selected as the subsystems replaced in the system expansion (Margallo et al., 2014a). According to the Think step databases, the contribution of each energy source to the Spanish grid mix (compiled in Appendix B) is valid for years 2013-2019.

On the other hand, the combustion process of natural gas is considered part of the life cycle of FA carbonation. Thus, the environmental impacts related to the generated electricity (the secondary system function) are subtracted from the system, allowing the comparison between FA carbonation and FA stabilisation. Since the combustion of natural gas can be coupled to a turbine that generates electricity, it is assumed that i) the generated electricity substitutes the same amount of electricity from the Spanish grid mix (average value) and ii)

the generated electricity substituted the energy obtained from natural combustion. A strong connection exists between the selected LCA approach and the choice of how to treat co-products. There are two LCA approaches: attributional LCA (ALCA) and consequential LCA (CLCA). In this study, ALCA perspective was selected since it describes physical (pollution and resource) flows within a chosen system (Thomassen et al., 2008), assuming that the analysed system does not affect its environment (Bala et al., 2015).

2.4 Life cycle inventory and modelling

The life cycle inventory is one of the most effort-consuming steps, and consists on the collection and interpretation of the data necessary for the environmental assessment of the observed system (Iannone et al., 2014). In the case of FA stabilisation, input and output data are based on a bibliographic source. In both scenarios the wastewater treatment plant was modelled assuming a primary and secondary treatment, and the inert landfill was modelled based on the previous works of Margallo et al. (2014a). Secondary data were taken from the databases of Think step (Think step, 2017) and Ecoinvent (Ecoinvent, 2016).

Table 1. Life cycle inventory of scenario b: FA stabilisation

	Flows	Amount	Units	Data source
Inputs	Fly ashes	1.00	t FA	
	Cement	0.30	t / t FA	Biellen et al. (2014)
	Water	389	l/ t FA	Biellen et al. (2014)
Outputs	Stabilised ashes	1.69	t / t FA	Biellen et al. (2014)

To estimate the life cycle inventory of the carbonation process, we must take into account that this is a multi-stage process (Velts et al., 2011), based on a complex gas-liquid-solid reaction mechanism (Ji et al., 2018). Nonetheless, several studies suggest that the

dissolution and diffusion of CO_2 in the liquid phase is the slowest step (He et al., 2006), and therefore it controls the kinetics of the carbonation reaction (Pan et al., 2013; Pan et al., 2015). Hence, the proposed model considers that the CO_2 mass transfer rate to the liquid phase equals its reaction rate.

To estimate the $k_L a_{CO_2}$ coefficient (the product of the CO_2 mass transfer coefficient in water and the interfacial area of the system) the mass balance of the reacted CO_2 was formulated (Equation 1). The CO_2 mass transfer rate to the liquid phase (r_T) is proportional to the difference between the CO_2 equilibrium concentration (C_{eq}) and the CO_2 concentration in the liquid phase (C), as described in Equation 2. Since it is assumed that all the CO_2 that is transferred to the liquid phase reacts, the CO_2 concentration in the liquid phase at the reactor outlet is 0 mol·L^{-1}. The model considers a continuous stirred tank reactor with perfect mixing, in which the CO_2 concentration at the reactor outlet is the same as the CO_2 concentration in the liquid inside the reactor; i.e., $C = 0$ mol·L^{-1}. C_{eq} is calculated with Henry's Law (Equation 3), which states that C_{eq} is directly proportional to the CO_2 partial pressure in the gas phase.

$$F_{CO2} = r_T \cdot V_L \qquad \text{(Eq. 1)}$$

$$r_T = k_L a_{CO2} \cdot (C_{eq} - C) \qquad \text{(Eq. 2)}$$

$$C_{eq} = k_h \cdot P_T \cdot y \qquad \text{(Eq. 3)}$$

Solving these three equations with the values of Henry's constant k_h, the flowrate of CO_2 that is transferred to the liquid phase and reacts F_{CO2} (estimated from the experimental results), the parameters of the experimental setup (the volume of liquid in the reactor V_L, the gas pressure P_T and the molar fraction of CO_2 in the gas stream at the reactor outlet y), all of which are compiled in Appendix B, it was determined that the value of $k_L a_{CO_2}$ under

the experimental conditions is 0.00343 s^{-1}.

Mass transfer coefficients depend mainly on the geometry of the system, the properties of the fluids (CO_2 and water in this case), and their dynamics (Molga and Westerterp, 2013). Assuming that these characteristics and the interfacial area of the system remain constant, the estimated value of $k_L a_{CO_2}$ can be used to scale up the process.

Therefore, solving these equations for V_L, the volume of the carbonation reactor required to carbonate the FA generated yearly in Cantabria was calculated for different gas pressures and compositions, as shown in Appendix B. The system has been modeled for pressures from 1 to 5 bar and different percentages of CO_2 excess in the flue gas: 10, 50 and 100%.

The variation of these parameters was proposed to assess the sensitivity of the LCA results to the electricity consumption for agitation and compression purposes. Increasing the pressure of the gas stream and its CO_2 composition accelerates the kinetics of the process, which reduces the required reactor volume and thus the agitation power. On the contrary, raising the pressure of the gas stream increases the electricity consumed to compress the gas, whereas a higher CO_2 content in the gas stream decreases the gas flowrate and as a consequence, the compression energy. The equations and parameters needed to calculate the agitation and compression energy are detailed in Appendix B. Table 2 shows a summary of the life cycle inventory of carbonation for a pressure of 5 bar and 55 % excess of CO_2 in the flue gas.

Table 2. Life cycle inventory of carbonation (5 bar / 55 % excess of CO_2 in the flue gas)

	Flows	Amount	Units
Inputs	Fly ashes	1.00	t FA
	Energy		
	Compression	2.40E-02	kWh/ t FA
	Stirring	281	kWh/ t FA

	Natural gas	241	kg / t FA
	Water	6,000	l/ t FA
Outputs	Treated ashes	7.17	t / t FA

2.5 Life cycle impact assessment

The life cycle impact assessment stage was conducted with the LCA software GaBi 6 (Thinkstep, 2017). The impacts analysed in this study are divided in demand for natural resources (materials, water and energy) and environmental impacts (climate change, acidification, human toxicity and eutrophication). These impact categories have been chosen because i) quantifying the consumption of natural resources allows identifying strategies to minimise resource consumption, ii) climate change and acidification potential are pressing issues nowadays, iii) human toxicity potential takes into account the detrimental consequences of the processes on the ecosystem, which are the reason FA cannot be directly landfilled, and iv) eutrophication potential allows us to measure the impact on water resources.

For climate change, the IPCC method was applied (IPCC, 2013), whereas for human toxicity potential, acidification potential and eutrophication potential, the CML method (Guineé et al., 2001) was used. The demand for water, energy and resources was based on the environmental sustainability assessment (ESA) developed by Margallo et al. (2014c).

3. RESULTS AND DISCUSSION

In this section, the contribution of each subsystem to the studied impact categories is analysed and the LCA results of the three scenarios are compared. The LCA of the carbonation scenarios was performed considering the flue gas pressure and CO_2 excess that minimise the energy requirement, after assessing the influence of the operating parameters on the energy consumption.

Figure 3 depicts the climate change impacts for different values of pressure in the flue gas (from 1.0 to 5.0 bar) and percentages of excess CO_2 (10%, 55% and 100%). Each point in Figure 3 reflects a different reactor volume.

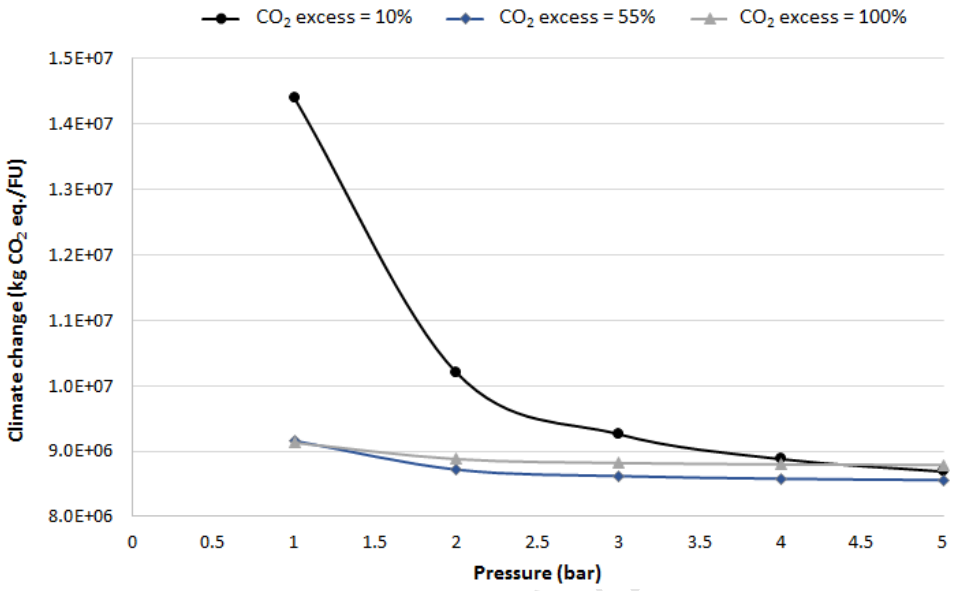


Figure 3. Climate change impact (kg CO_2 eq.) with the variation of pressure and excess CO_2 in the flue gas.

When the pressure increases, a higher equilibrium concentration is obtained and therefore, the required reactor dimensions are smaller, that is, diameter and height are reduced. The stirring power depends on the agitator diameter and this is conditioned by the reactor diameter, which is raised to the fifth (as shown in Equation A3 of Appendix B). Thus, as the flue gas pressure increases, less energy is required for agitation. The total energy of the carbonation process is equal to the sum of the energy required for compression and stirring, being the latter significantly higher than the former. As a consequence, the total energy consumption decreases as the flue gas pressure increases. Thus, the environmental impacts of the carbonation process decrease as the pressure increases (Margallo et al., 2018b).

As shown in Figure 3, the best conditions were achieved for a pressure of 5 bar, whereas a

similar impact (due to the similar required reactor sizes) was observed for the 55% and 100% CO_2 excess. For the 10% excess, the reactor dimensions are noticeably higher.

3.1 FA carbonation based on natural gas combustion (Scenarioa.1)

Taking the analysis of the operating conditions into account, a pressure of 5 bar and a 55% of CO_2 excess in the flue gas were selected to study the carbonation scenarios. Figure 4 evaluates the environmental impact categories for FA carbonation based on the use of natural gas as a CO_2 source.

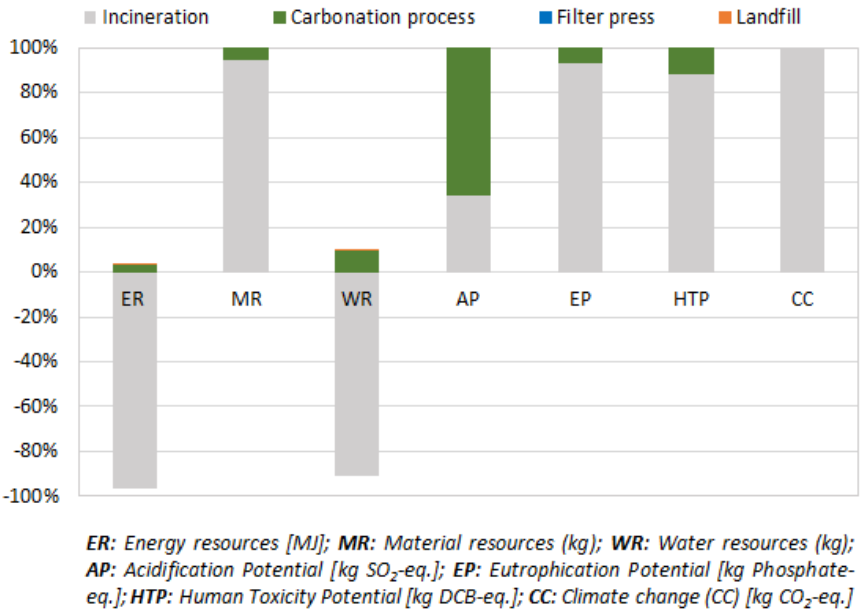


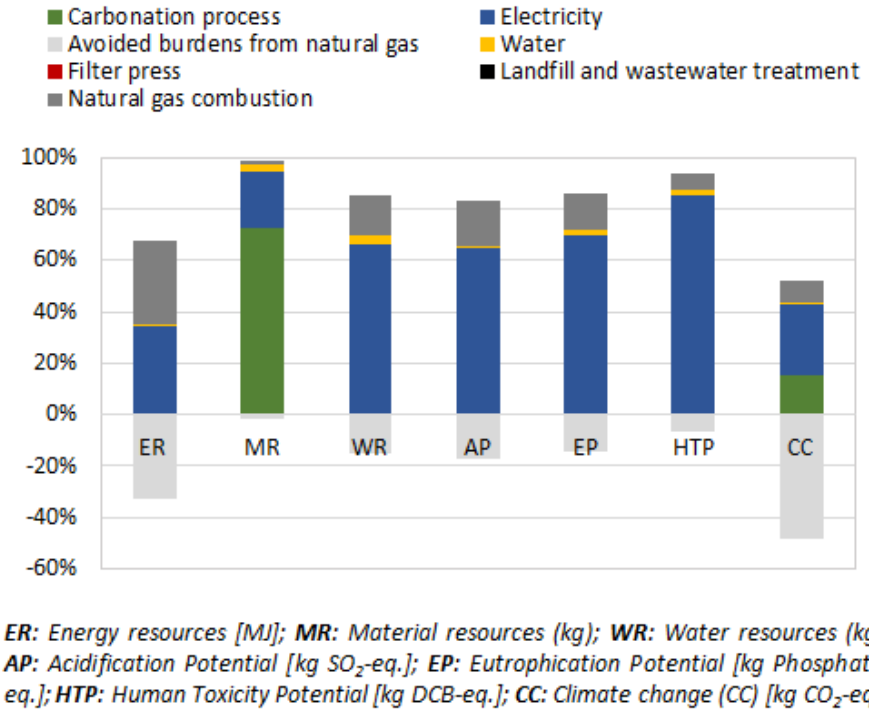
Figure 4. Contribution (%) of all the stages (incineration, carbonation, filter press, landfill and wastewater treatment) to the impact categories for FA carbonation based on the combustion of natural gas.

The results include the impacts from the incineration process, carbonation, filter press, landfilling and wastewater treatment. MSW combustion has a contribution from 88.5% to 99.8% in the categories of materials resources (MR), eutrophication potential (EP), human toxicity potential (HTP) and climate change (CC). This is due to the emission of

greenhouse gases, especially CO_2 (in CC), and toxic substances, mainly dioxins and furans (in HTP), as well as the release of ammonia, phosphorus, phosphate and nitrogen to water and nitrogen oxides and nitrous oxide to the air compartment (in EP). On the other hand, the WtE plant produces energy and steam, which, in accordance with the applied system boundary expansion method, replace the equivalent amount of energy from the Spanish grid mix and the consumption of water required for the generation of energy from other sources. Since the amount of energy and water required in this scenario is lower than the amount of energy and water being displaced, negative values are obtained for the MR and ER categories. The carbonation process has the highest influence, c.a. 66.6% in the category of acidification potential (AP), due to the emission of acidifying substances, such as SO_2, HCl and HF, in the generation and consumption of energy. This process is also significant, but to a lesser extent, in the consumption of natural resources, and in the categories of EP and HTP, ranging from 5.5% to 11.5%.

The impact of the filter press, the landfilling of ash, and the wastewater treatment is negligible in comparison to the contribution of the combustion process. If the combustion process is excluded from the system boundaries (Figure 5), the consumption of electricity in the carbonation produces the highest impacts, followed by the combustion of natural gas for CO_2 generation.

In particular, the consumption of energy has impacts from 22.0% to 99.0% in most of the categories, whereas the carbonation process has a great influence in the consumption of materials. The contributions of the landfill and the wastewater treatment are below 1% and thus, these impacts can be neglected. The negative values are associated with the avoided burden of natural gas, whose combustion produces energy that is an extra function of the system.



***ER:** Energy resources [MJ]; **MR:** Material resources (kg); **WR:** Water resources (kg); **AP:** Acidification Potential [kg SO_2-eq.]; **EP:** Eutrophication Potential [kg Phosphate-eq.]; **HTP:** Human Toxicity Potential [kg DCB-eq.]; **CC:** Climate change (CC) [kg CO_2-eq.]*

Figure 5. Evaluation of FA carbonation based on the combustion of natural gas excluding the incineration process.

On the other hand, the allocation procedure has an influence on the results; Figure 6 compares the environmental performance of the system considering two alternatives: that the energy generated in the natural gas combustion process substitutes electricity from the Spanish grid mix or electricity derived from the combustion of natural gas. The allocation based on natural gas has a greater impact than the substitution of electricity from the grid mix, therefore a larger environmental benefit is achieved with the second allocation method in most of the impact categories.

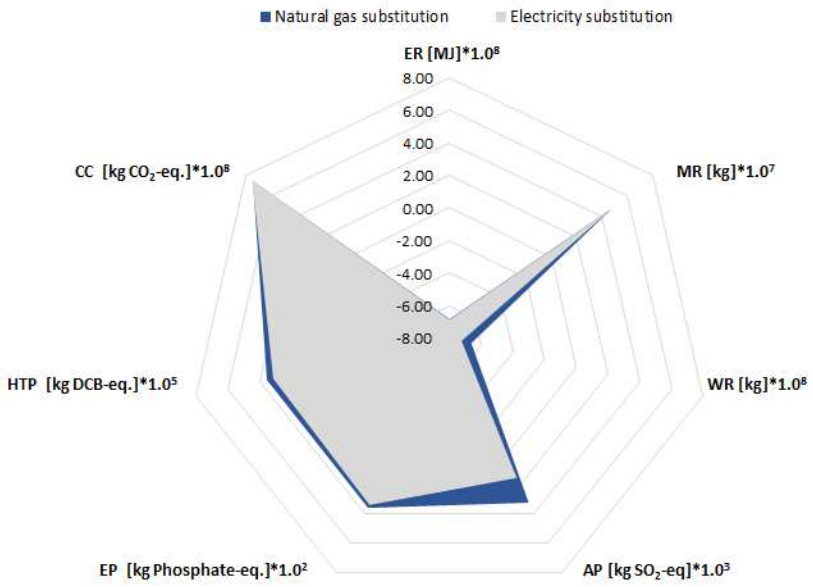


Figure 6. Comparison of FA carbonation based on the combustion of natural gas using different allocation strategies for energy production: substitution of electricity from the combustion of natural gas and electricity from the Spanish grid mix.

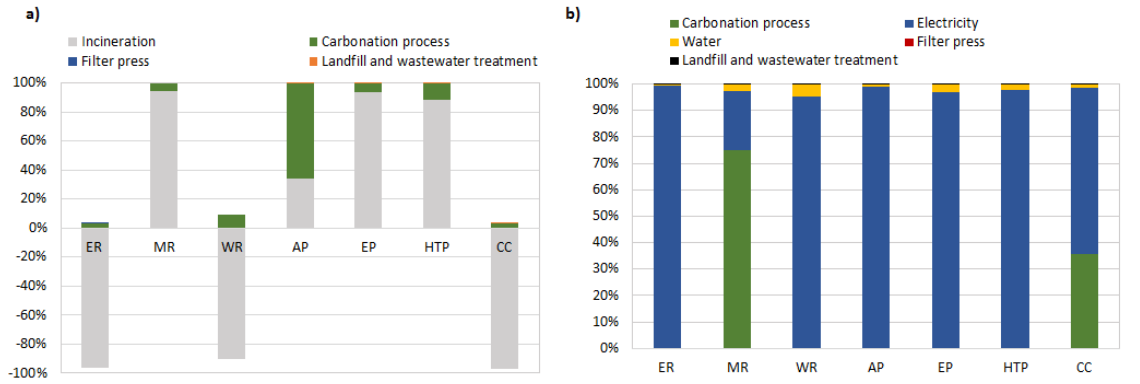
3.2 FA carbonation based on the use of WtE flue gases (Scenario a.2)

Figure 7.a shows the contribution of incineration, carbonation, filter press, landfilling and wastewater treatment to the studied impact categories when the flue gases from the WtE plant are used as a CO_2 source for the selected operating conditions of 5 bar and 55% CO_2 excess in the flue gas.

As in Scenario a.1, MSW combustion has the highest contribution to most of the categories in Scenario a.2, only in the category of acidification potential the carbonation process provides 65.5% of the impact due to the emission of acidifying substances in the generation and consumption of electricity. Likewise, since the energy generated in the incineration process is assumed to replace other sources of energy, negative ER and MR values are obtained in Scenario 2. The main difference regarding the source of CO_2 is observed in the

category of climate change; Scenario a2 has lower CC impacts than Scenario a1.

If the analysis excludes MSW combustion, the consumption of electricity in the carbonation produces the highest impacts in most of the categories (Figure 7b), with contributions between 22.2% in MR to 99.3% in ER. Additionally, the carbonation process presented a notable contribution to the CC category due to the emission of CO_2.



***ER:** Energy resources [MJ]; **MR:** Material resources (kg); **WR:** Water resources (kg); AP: Acidification Potential [kg SO_2-eq.]; **EP:** Eutrophication Potential [kg Phosphate-eq.]; **HTP:** Human Toxicity Potential [kg DCB-eq.]; **CC:** Climate change (CC) [kg CO_2-eq.]*

Figure 7. Contribution (%) of a) all the stages (incineration, carbonation, filter press, landfill and wastewater treatment) and b) all the stages excluding the incineration process for FA carbonation based use of the flue gases from MSW combustion.

3.3 Stabilisation process (Scenario b)

Figure 8a shows that in the stabilisation scenario, the largest contributor to all the impact categories except AP is the incineration process. In the AP category, the use of water and the production and consumption of cement is responsible for 63.8% of the total impact.

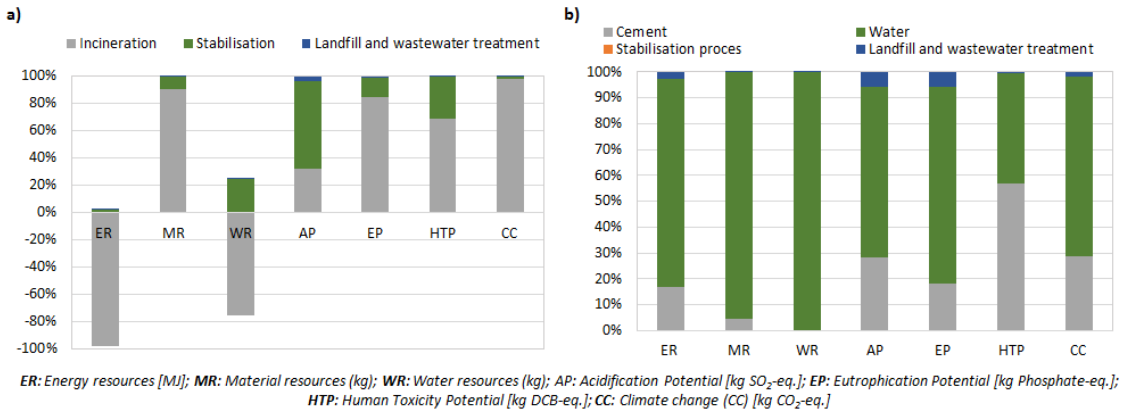


Figure 8. Contribution (%) of a) all the stages (incineration, stabilisation, landfill and wastewater treatment) and b) all the stages excluding the incineration process for FA stabilisation.

Figure 8b analyses the results excluding combustion. FA stabilisation has three stages (stabilisation, cement production and water treatment). The stabilisation stage does not contribute to any of the impact categories, because it is assumed that the process is based solely on the mixing of the materials, and energy is not required. The demand for water in the stabilisation scenario has a contribution of 42-99.9% to the studied categories, with the highest contribution in the consumption of natural resources and eutrophication categories. Cement production, which is a highly energy demanding process, has the greatest impact in human toxicity (57%), and a contribution of around 28% to the climate change and acidification impact categories.

3.4 Comparison of FA treatments (Scenario b)

Figure 9.a compares the results (which have been normalised multiplying them by the negative powers of 10 shown in the figure) of the three scenarios under study: carbonation based on the combustion of natural gas (Scenario. a.1), carbonation based on the use of flue gases from the WtE plant (Scenario a.2) and stabilisation (Scenario b). Figure 9.b only

includes Scenario a.1 and Scenario a.2.

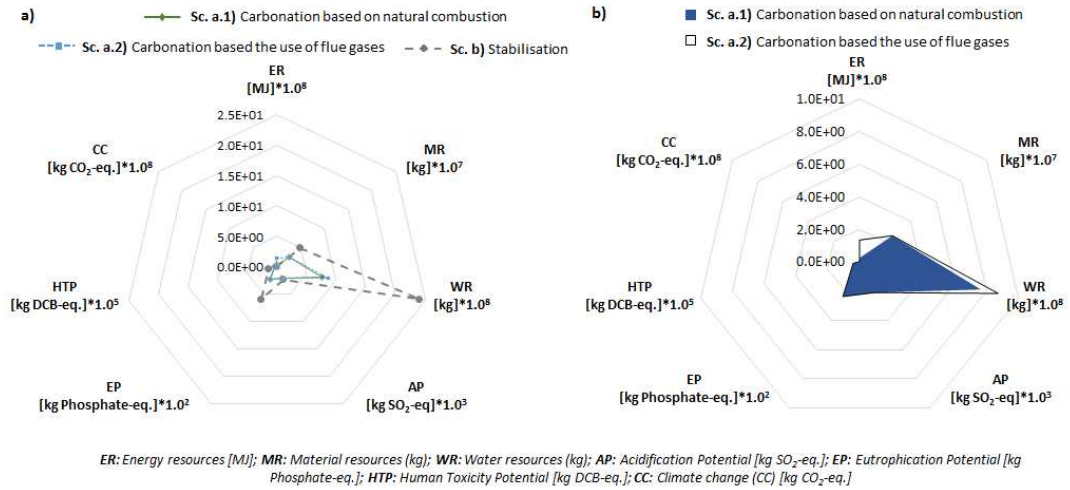


Figure 9. Normalised LCA results. a) Comparison of FA carbonation (Scenario a.1 and a.2) and stabilisation (Scenario b), and b) comparison of carbonation scenarios a.1 and a.2.

It can be clearly seen that stabilisation has higher impacts than carbonation in most of the categories mainly because of the reduction of lixiviation and the CO_2 capture in the ash. Only in the ER category, carbonation has greater impact than stabilisation due to the electricity consumption for agitation and compression purposes.

The source of CO_2 used for the carbonation process also had a significant influence on the results (Figure 9b). Scenario a.1 and Scenario a.2 showed similar values for the categories of MR, EP and HTP; whereas for ER and WR, the use of CO_2 from the MSW combustion presented a higher impact. This happens because the combustion of natural gas, which generates the CO_2 that is subsequently used in the carbonation process, also produces energy that is assumed to displace electricity from the Spanish grid mix. This avoided burden has a significant influence on the consumption of energy and water; that is why these impact categories are meaningfully reduced.

4. CONCLUSIONS

The accelerated carbonation of the FA generated in the incineration of MSW is an innovative technology with the potential to capture CO_2 and simultaneously immobilise the pollutants contained in the FA. Nevertheless, prior to the implementation of the accelerated carbonation process within a MSW management system, its sustainability implications must be assessed.

The analysis of the operating conditions of the carbonation process revealed that the low solubility of CO_2 in water is the main factor that hinders the large-scale implementation of the process. Large reactor volumes, associated with a noteworthy energy consumption in the stirring process, are required to overcome the low solubility of CO_2. Since the energy required for stirring and the agitator diameter follow an exponential relationship, placing several smaller reactors in parallel could be an alternative to a single large reactor that should be studied in detail. On the other hand, increasing the pressure of the gas stream that is fed to the reactor accelerates the kinetics of the reaction and consequently decreases the reactor volume, at the expense of on an increase in the compression energy. It was found that the overall energy consumption of the carbonation process is minimised at the upper bound of the range of flue gas pressures studied.

The LCA results, subject to the assumptions made in the allocation procedure, revealed that FA carbonation had lower impacts than stabilisation in the studied impact categories, except for the consumption of energy resources, consumption of raw materials, acidification, eutrophication, human toxicity, and climate change.

Therefore, if further research succeeds in minimising the energy consumption of the carbonation process, it could become a more sustainable alternative to stabilisation,

allowing waste managers to use one waste stream (the flue gas generated in the incineration process)to treat FA, another waste stream generated in the same process.

Nonetheless, the feasibility of the carbonation process cannot be established until an economic analysis guarantees its profitability; coupling economic metrics to the developed LCA model of the system will be essential to help waste managers make informed decisions.

5. ACKNOWLEDGMENTS

The authors are grateful for the funding of the project "Estudio de la estabilización de cenizas volantes de incineración mediante carbonatación (SADER)". Moreover, the authors wish to extend their acknowledgment to all people involved in the LIFE FENIX Project, which this work was a part of, as well as the European LIFE Financing Programme.

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- LCA performed to evaluate the environmental performance of fly ash treatments
- Fly ash stabilization is a less favourable alternative compared to carbonation
- Energy consumption in carbonation decreases as pressure of the flue gas increases
- Best operational conditions of carbonation for 3 to 5 bar and a 55% excess of CO_2
- The CO_2 source in the carbonation has a high influence in the environmental impacts