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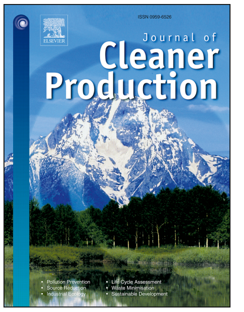

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Life-cycle assessment as a tool to evaluate the environmental impact of hot-dip galvanisation

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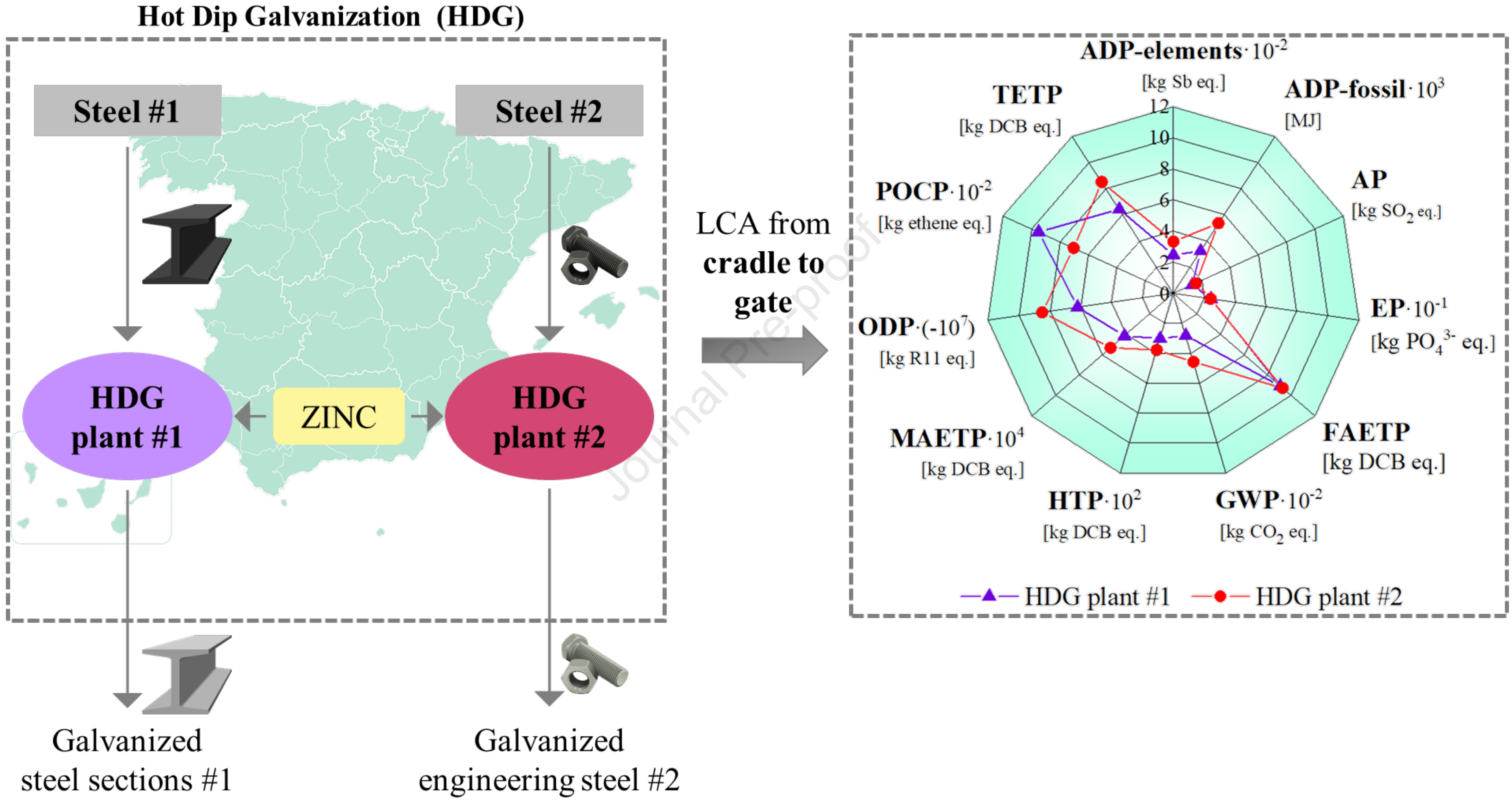

Hot Dip Galvanization (HDG)
Steel #1
Steel #2
HDG plant #1
ZINC
HDG plant #2
Galvanized steel sections #1
Galvanized engineering steel #2
LCA from cradle to gate
ADP-elements·10^{-2} [kg Sb eq.]
ADP-fossil·10^{3} [MJ]
AP [kg SO_2 eq.]
EP·10^{-1} [kg PO_4^{3-} eq.]
FAETP [kg DCB eq.]
GWP·10^{-2} [kg CO_2 eq.]
HTP·10^{2} [kg DCB eq.]
MAETP·10^{4} [kg DCB eq.]
ODP·(-10^{7}) [kg R11 eq.]
POCP·10^{-2} [kg ethene eq.]
TETP [kg DCB eq.]
0
2
4
6
8
10
12
HDG plant #1
HDG plant #2

ABSTRACT

Hot-dip galvanisation (HDG) is the method most commonly used to protect steel surfaces from corrosion. However, HDG involves very intensive consumption of energy and resources. This paper aims to evaluate the environmental performance and hotspots in the Spanish HDG sector using cradle-to-gate life cycle assessment (LCA). Two Spanish HDG industrial plants, with different galvanisation capacities and manufacturing processes, were selected for the case study. The LCA revealed that the consumption of energy, fuels and nonrenewable resources were the most relevant environmental burdens at both plants. Steel was the main contributor, as it had the greatest influence on the plants' environmental profiles. The consumption of primary zinc and natural gas, used to dry and heat the molten zinc bath, also contributed to the impact of the HDG plants. This work proposes a normalisation to compare the Spanish sector against a European reference based on the average of 66 companies in 14 countries. The impacts of the Spanish HDG plants being studied were generally below the EGGA values, although the results are strongly influenced by the type of steel and the degree of material reuse implemented in the steel manufacturing process. The study lays the foundations for improvements in the resource efficiency and productivity of galvanisation plants. We propose alternatives such as the use of secondary zinc or modifications that will extend the lifespan of pickling baths, which would contribute to a more sustainable use of resources in the galvanising industry.

Keywords: galvanized steel, hot-dip galvanization (HDG), life cycle assessment (LCA), normalization, zinc, spent pickling acids.

Abbreviations

ADP-elements: Abiotic depletion of elements

ADP-fossil: Abiotic depletion of fossil fuels

AGA: American Galvanizers Association

AP: Acidification potential

ATEG: Asociación Técnica Española de Galvanización

BF: Blast furnace

BOF: Basic oxygen furnace

EAF: Electric arc furnace

EGGA: European General Galvanizer Association

EP: Eutrophication potential

EPD: Environmental product declaration

FAETP: Freshwater aquatic ecotoxicity potential

FU: Functional unit

GPD: Gross domestic product

GWP: Global warming potential

HDG: Hot-dip galvanising

HTP: Human toxicity potential

LCA: Life-cycle assessment

LCCA: Life-cycle cost assessment

LCI: Life-cycle inventory

LCIA: Life-cycle impact assessment

MAETP: Marine aquatic ecotoxicity potential

ODP: Ozone layer depletion potential

PAH: Polycyclic aromatic hydrocarbons

PCR: Product category rules

POCP: Photochemical ozone creation potential

SHG: Special high-grade

SPA: Spent pickling acid

TETP: Terrestrial ecotoxicity potential

1. INTRODUCTION

Steel is the primary material used to manufacture vehicles, household appliances and industrial machinery (Shibli et al., 2015). Global steel production was 1,869.9 million tonnes (Mt) in 2019, with China accounting for 53.3% of all production and Europe contributing 16.0% (Worldsteel Association, 2020). However, due to corrosion, steel has a limited lifetime. The costs associated with steel corrosion have been estimated to correspond to 4.0% of the gross domestic product (GPD) in countries such as Japan and USA (Woolley, 2008). Zinc coating is one of the most widely used corrosion-prevention measures because the zinc acts as an anode and corrodes before the underlying steel. There are several zinc coating methods available for protecting steel components, including hot-dip galvanising (HDG), electroplating, metallising (zinc spraying), mechanical plating and zinc-rich painting (Urtiaga et al., 2010). Of the various zinc coating techniques, HDG offers a combination of superior functional properties and low cost (Shibli et al., 2015). In HDG, a protective zinc layer is formed by immersing steel pieces in a bath of molten zinc, thereby creating a series of zinc–iron alloy layers metallurgically bonded to the steel. The HDG process typically produces zinc coatings that are 45–200 µm thick (EGGA, 2020). Therefore, the underlying steel is protected by two mechanisms: a physical barrier and electrochemical protection (Hegyi et al., 2017). However, despite the advantages of galvanisation, the corrosion protection process entails a considerable increase in production costs and sale price of the final product. In this context, Rossi et al. (2017) carried out a comparative life-cycle cost assessment (LCCA) for a steel girder bridge protected with either anticorrosion paint or through HDG. The authors concluded that the HDG process delivers an economic advantage when it is compared to painting, using socio-economic indicators such as the Net Present Value (NPV). In addition to the economic and technical benefits of galvanisation, we need to consider the environmental performance of the product, as HDG is a very intensive process in terms of primary zinc and energy consumption (Kong and White, 2010). Approximately 50% of the zinc produced in the world is used to galvanise steel pieces, resulting in a correlation between steel and zinc production cycles (Watari et al., 2020). The European batch HDG industry uses, on average, 400,000 tonnes of zinc to coat 5 million tonnes of steel each year (0.08 t of zinc/t of steel) (ATEG, 2020), and most of the zinc consumed comes from primary sources. Yet primary zinc production is a highly intensive process with respect to resource and energy usage (Ng et al., 2016), which are consumed at even higher rates than in steel production (Norgate et al., 2007). Furthermore, the generation and disposal of harmful residues produced during HDG, in the form of spent pickling acids (SPAs) loaded with metals, also increase the environmental burden of galvanisation (Culcasi et al., 2019; Ortiz et al., 2004). Landfills have been the predominant alternative for the disposal of municipal solid waste as well as industrial waste worldwide (Camba et al., 2014). However, although waste is kept in a long-term safe and strict regulations control landfilling, it is a potential source of pollution and hazardous substances (Diban et al., 2011).

Life-cycle assessment (LCA) is considered an accurate method for evaluating the potential impact of hot-dip galvanisation, so that its results may be used to propose alternatives to improve its environmental performance (García et al., 2013). Several authors have applied LCA to the steel industry. Norgate et al. (2007) compared the environmental performance of various metal production processes, including steel production through blast furnace (BF) and basic oxygen furnace (BOF) techniques. Tongpool et al. (2010) evaluated the environmental impact of steel products such as steel slab, hot-rolled and cold-rolled steel, hot-dip galvanised and electrogalvanised steel. Hao et al. (2012) identified the stages of the steel production with the highest environmental burdens using different impact assessment methods, namely EDIP, CML 2001 and Eco-indicator 99. However, very few studies have assessed the environmental impact and resource usage of the HDG process. In this regard, De Benedetti et al. (2003) reported objective and reliable information that can be used to compare different anticorrosion alternatives, such as hot-dip galvanising, replacing steel with stainless-steel or painting, in terms of global warming potential (GWP) and acidification potential (AP). The authors concluded that the production of the steel and zinc required to produce galvanised steel were the main contributors to the GWP and AP indicators. Additionally, Hernández-Betancur et al. (2019) developed a holistic approach that includes uncertainty, taking into account the three dimensions of sustainable development, to determine the capability of HDG to achieve a satisfactory level of sustainability. Results showed that the stages of pickling and fluxing are critical, and they are not expected to improve without adjustments in the HDG process. Moreover, the European General Galvanizers Association (EGGA) and the American Galvanizers Association (AGA) have both attempted to develop tools to assess the sustainability of HDG. The EGGA conducted a sectoral Environmental Product Declaration (EPD) for hot-dip galvanised steel items (EGGA, 2016) based on the Product Category Rules (PCR) 2011:16 “Corrosion protection of fabricated steel products” (The International EPD System, 2017). On the other hand, the AGA assessed the environmental performance of hot-dip galvanised steel (AGA and IZA, 2017) through an LCA and an EPD based on the North American PCR for Designated Steel Construction Products (SCS Global Services, 2015). Both associations considered the production, use and end-of-life phases, and the benefits associated with recycling galvanised steel. While these works are informative technical reports, they do not include a complete discussion or any advances in the LCA methodology.

Considering this background, the present work analyses the environmental impacts of the HDG process in Spain from cradle to gate, including the steel production step. We present some novel aspects in this regard. This is the first paper that provides a complete and thorough LCA study of the HDG process from an environmental point of view. Previously De Benedetti et al. (2003) evaluated the energy requirements of galvanising and impact categories such as GWP and AP were analysed briefly and to a lesser extent than in the present study. Besides, this is the first work that quantifies the contribution of each stage of the HDG process to each impact category. Although the associations EGGA and AGA and De Benedetti et al. (2003) assessed the environmental burdens of the HDG process, those studies did not evaluate impact categories related to the toxicity. Therefore, this work expands the environmental results by including the impact categories Freshwater Aquatic Ecotoxicity Potential (FAETP), Human Toxicity Potential (HTP), Marine Aquatic Ecotoxicity Potential (MAETP) and Terrestrial Ecotoxicity Potential (TETP). Despite previous studies determined that steel and primary zinc productions represent the main contributions to the environmental impacts of the HDG process, they did not present an in-depth background evaluation of both manufacturing processes. This work analyses in detail the causes behind the environmental impacts of both productions. After identifying the environmental impacts, we propose alternatives to improve resource usage and the environmental performance of HDG. Furthermore, this work constitutes the first LCA of the Spanish HDG process, based on two HDG plants located in Spain. Both installations are representative of the sector due to their differences in the production capacity (1:15) and characteristics of the process associated with the final application of the steel items. The steel items that are galvanised in each HDG plant are different and this allows an environmental comparison based on the type of steel product. Finally, we propose a new external normalisation that allows an environmental assessment of Spanish HDG plants compared to a sample representative of the European industry provided by EGGA. The normalisation procedure is not novel, but the threshold values are being used for the first time and can be an important aid in the normalisation process, as they provide an overview of the environmental performance of the HDG process at the European level. This procedure simplifies results interpretation and facilitates decision-making with regard to tracking the progress made towards environmental sustainability, improving resource and energy efficiency, and promoting a circular economy.

2. LIFE CYCLE ASSESSMENT METHODOLOGY

LCA is a standardised tool for compiling and evaluating the inputs, outputs and potential environmental impact of a given product system throughout its life cycle, that is, from raw material extraction to end-of-life (ISO, 2006a). The tool employs an iterative and holistic technique that can be used to support decision-making by identifying the hotspots of a product, process or service (Rebitzer et al., 2004). According to ISO Standards 14040 and 14044 (ISO, 2006a, 2006b), an LCA comprises four phases: goal and scope definition, life-cycle inventory (LCI), life-cycle impact assessment (LCIA) and interpretation of the results.

2.1 Goal and scope definition

The goal and scope definition provides a description of the product system in terms of system boundaries and in relation to a functional unit (FU) (Rebitzer et al., 2004). Thus, the choices made at this stage will influence the entire study (Pierucci et al., 2017). The aim of this work was to evaluate the resource usage and environmental impacts of the HDG process based on a cradle-to-gate approach.

2.1.1 Function and functional unit

The system's main function is to protect steel components from corrosion, hence we selected one tonne of galvanised steel as the FU. This selection was based on a previous publication involving an LCA of primary zinc production (Van Genderen et al., 2016). In addition, one of the HDG plants in our comparison featured the extra function of producing thermal energy and electricity in a cogeneration unit. The handling procedure for this multifunctional system is described in the Allocations section.

2.1.2 System description and boundaries

Fig. 1 shows the system boundaries of both HDG plants. The study included the extraction of raw materials, their transport to the HDG plants, the HDG process, the transport of residues to waste management facilities, waste and wastewater treatment, and the final disposal. We also included steel production in order to quantify its contribution to the whole system. The infrastructures involved in the manufacturing process were not considered because their impacts are insignificant considering the long lifetime of the industrial installations (Dominguez et al., 2018). The use, maintenance and end-of-life of galvanised steel are not included in the scope of this study.

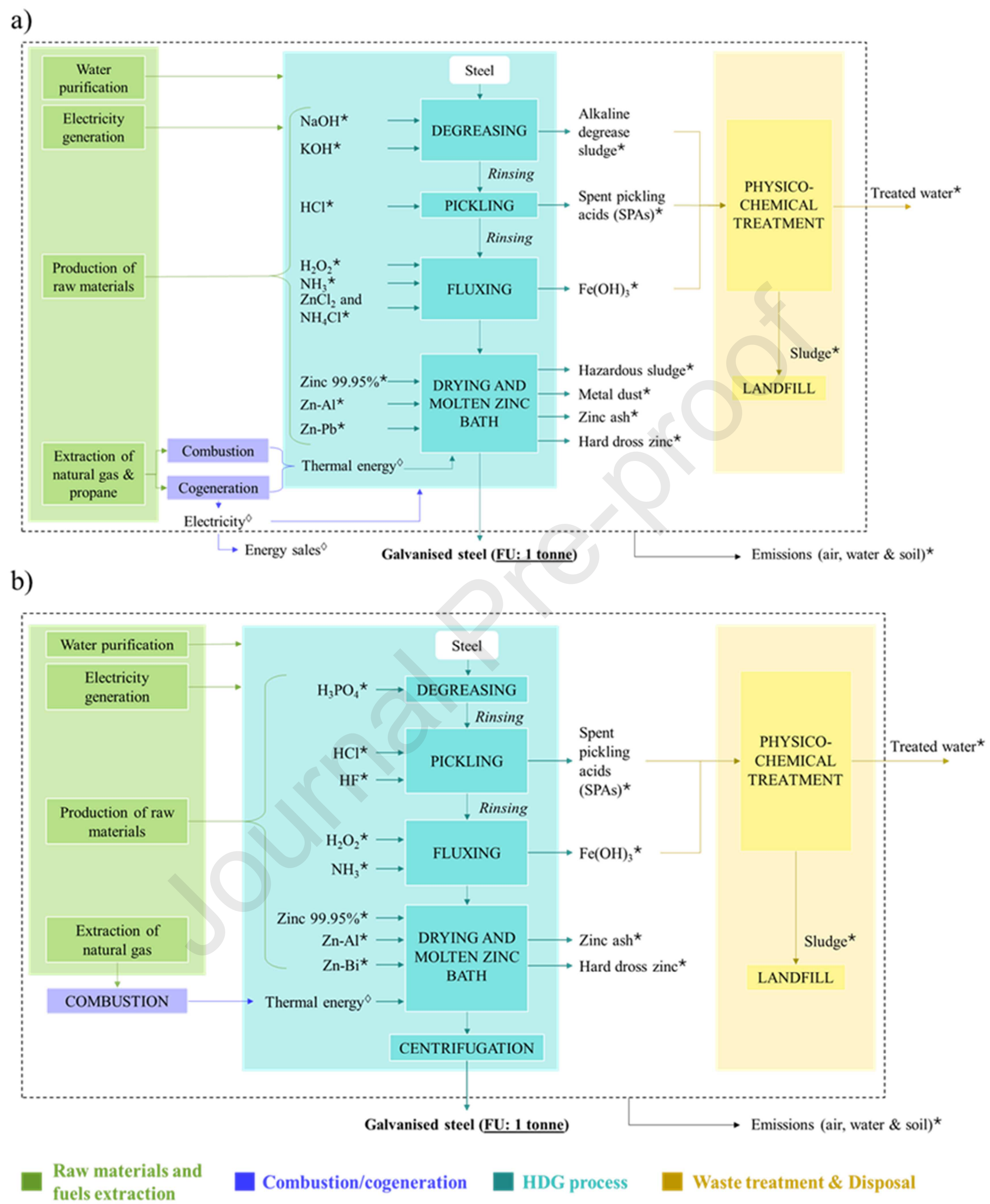

Fig. 1. System boundaries of a) HDG plant #1 and b) HDG plant #2 with "★" and "◊" as unit symbols of mass (kg) and energy (MJ).

Two HDG plants were analysed in order to represent the galvanising sector in Spain. The plants are located 600 km apart and typically serve different markets and clients. HDG plant #1 is engaged mostly in the production of steel profiles and sections for the construction sector, whereas HDG plant #2 deals with engineering steel used in the manufacture of wind turbines, engine components, and so on. Moreover, the production capacity of HDG plant #1 is 15 times greater than that of HDG plant #2.

Both plants implement the same main stages in the galvanisation process: degreasing, pickling, fluxing, drying and molten zinc bath; although with some differences in the operation mode and materials used. HDG plant #2 also incorporates a centrifugation stage to remove excess zinc (see **Fig. 1b**). First, the steel piece is degreased with either an alkaline or acidic agent to remove any organic impurities from the steel surface (Shibli et al., 2015). HDG plant #1 employs an alkaline degreasing bath consisting of an aqueous solution of sodium hydroxide (NaOH) and potassium hydroxide (KOH). HDG plant #2, for its part, operates an acidic degreasing stage with phosphoric acid (H_3PO_4). In the following pickling stage, iron oxides are removed from the surface of the steel pieces. Both HDG facilities also use the pickling bath to remove zinc from reprocessed galvanised steel components. When the pickling rate is very low, part of the exhausted acid is replenished with 33% (v/v) hydrochloric acid (HCl) to extend the lifespan of the pickling bath (Culcasi et al., 2019). This amount of HCl used for the replenishment of the pickling bath is also normalised to the FU. Fluxing then prepares the surface to ensure correct contact between the steel and molten zinc. Hydrogen peroxide (H_2O_2) is added to oxidise any iron (II) to iron (III) and flux bath pH is adjusted to approximately 4.5 with ammonia to precipitate iron (III) hydroxide. Fluxing also protects the steel surface from oxidation by immersing the piece in a zinc chloride ($ZnCl_2$) and ammonium chloride (NH_4Cl) solution. Both salts are added at HDG plant #1, but HDG plant #2 eliminates this addition by taking advantage of the zinc released from reprocessed steel items and the ammonia used to adjust the pH. The drying stage uses thermal energy obtained from natural gas combustion, which is the main energy vector in both galvanising plants, to remove any moisture from the steel components. In addition, HDG plant #1 incorporates a cogeneration unit that produces thermal energy and electricity from the combustion of natural gas (see **Fig. 1a**). Propane is used for welding and the metallisation of steel pieces at HDG plant #1. Finally, steel items are immersed in a molten zinc bath at approximately 450 °C that makes the energy consumption of this stage the greatest by far (Kong and White, 2010). Special high-grade (SHG), 99.995% purity, zinc is used to give the steel a zinc alloy corrosion protection layer. Several alloying additives are incorporated: zinc-aluminium (Zn-Al) and zinc-lead (Zn-Pb) at HDG plant #1, and both Zn-Al and zinc-bismuth (Zn-Bi) at HDG plant #2. Aluminium forms an alumina (Al_2O_3) layer that prevents oxidation, while lead and bismuth reduce the surface tension of the molten zinc and therefore improve its castability (Kania et al., 2020).

2.1.3 Allocations

The main function of both HDG plants studied here is to protect steel items against corrosion. Moreover, HDG plant #1 has a cogeneration unit that produces electricity, which is partly consumed onsite at the plant, while the rest is sold to the electricity grid. The electricity sales represent an extra function involved in the process. Consequently, HDG plant #1 is defined as a multioutput process. Based on ISO 14040 guidelines (ISO, 2006a), in such multifunctional systems, the environmental burdens must be divided into different functions through system division or expansion, avoiding the use of causality and noncausality allocation. Therefore, the environmental performance of HDG plant #1 and

HDG plant #2 should be compared on the same footing, that is, based on the same function without incorporating extra functions. Accordingly, we applied system expansion to credit the additional electricity produced through cogeneration at HDG plant #1. The energy credits replace electricity from the Spanish national grid. However, when comparing the two HDG plants, we removed the energy credits to focus on the system's main function.

2.1.4 Cut-off criteria

The cut-off criteria identify the amount of material, energy flow or environmental significance associated with unit processes or product systems that should be excluded from the study (ISO, 2006a). This study established that each excluded material flow must not exceed 1% of the mass of each unit process and the sum of all excluded material flows in the system must not exceed 5.0% of the total mass flux. Furthermore, we considered all process energy inputs, as the HDG process is an energy-intensive process. Considering all these criteria, the production of $ZnCl_2$ and NH_4Cl salts (used in the fluxing stage) and zinc alloys (added to the molten zinc bath) were excluded from the study because they account for less than 1% of the total system input and output.

2.2 Life-cycle inventory

LCI is one of the most laborious steps and consists of data collection, calculation and procedures to quantify the relevant product system inputs and outputs (Iosif et al., 2009). In the present work, the primary data are based on questionnaires the companies completed with respect to 2017. For secondary data, we used Ecoinvent and Thinkstep databases (Ecoinvent, 2020; Sphera, 2020). Table S2.1 includes all the processes employed in each database. **Table 1** shows the life-cycle inventory for both plants in 2017. The data correspond to the functional unit considering that the production of HDG plants #1 and #2 was $51 \cdot 10^3$ and $3.5 \cdot 10^3$ tonnes of galvanised steel, respectively.

Table 1. Life-cycle inventory per FU (1 tonne of galvanised steel) of HDG plants #1 and #2 in 2017.

Input	**Units**	**HDG plant #1**	**HDG plant #2**
NaOH	kg/FU	0.23	-
KOH	kg/FU	0.06	-
H_3PO_4	kg/FU	-	0.22
HCl	kg/FU	25.4	9.1
HF	kg/FU	-	0.09
Inhibitor	kg/FU	-	0.036
Foaming	kg/FU	-	0.037
$ZnCl_2$	kg/FU	0.31	-
NH_4Cl	kg/FU	0.04	-
$ZnCl_2$ and NH_4Cl	kg/FU	0.55	-
H_2O_2	kg/FU	2.60	0.27
NH_3 24%	kg/FU	0.58	0.51
Natural gas (cogeneration)	kWh/FU	494	-
Natural gas (combustion)	kWh/FU	208	804
Propane	kg/FU	0.025	-
Zn-Pb	kg/FU	2.86	
Zn	kg/FU	58.9	79.2
Zn-Al	kg/FU	0.60	0.29
Zn-Bi	kg/FU	-	0.71
Water	kg/FU	233	222
Wire	kg/FU	1.63	-
Wood	kg/FU	2.87	-
Pallets	kg/FU	0.98	-
Electricity	kWh/FU	0.58	77.4
Output	**Units**	**HDG plant #1**	**HDG plant #2**
Alkaline degrease sludge	kg/FU	0.89	-
Spent acid	kg/FU	36.3	14.8
Soil containing hazardous substances	kg/FU	0.14	-
Metal dust	kg/FU	0.11	-
Sludge $Fe(OH)_3$	kg/FU	1.1	3.4
Zinc ash	kg/FU	6.5	28.2
Zinc dross	kg/FU	3.9	35.7
Water with HC	kg/FU	0.32	-
Scrap	kg/FU	3.0	-
Wood	kg/FU	1.15	-

Industrial waste	kg/FU	0.48	1.3
Contaminated materials	kg/FU	0.10	-

Steel production levels for HDG plants #1 and #2 is based on World Steel data from the Thinkstep database and is represented by "steel sections" and "engineering steel" respectively (World Steel, 2017). According to the Thinkstep database, engineering steel is processed in an electric arc furnace (EAF), while steel sections are manufactured through a combination of the BOF and EAF techniques. EAF steel production is reportedly less energy-intensive than BOF (Yellishetty et al., 2011). Besides, the EAF process is less intensive in terms of resource consumption and carbon dioxide (CO_2) emissions, since it employs scrap to produce steel, while the BOF process uses iron ore as its main input.

2.3 Life-cycle impact assessment

2.3.1 Classification and characterisation

We conducted the LCIA using the LCA software GaBi 9 (Sphera, 2020), with CML 2001 as the impact assessment method, which is a midpoint or problem-oriented method developed by a group of scientists under the supervision of the Institute of Environmental Sciences (CML) at Leiden University, The Netherlands. The problem-oriented approach uses values at the beginning or middle of the environmental mechanism, providing a detailed and transparent picture of the ecological impacts. The set of impact categories (**Table 2)** was selected in accordance with the seven CML impact categories used by the EGGA in its EPD (EGGA, 2016). Furthermore, we also added other impact categories to provide a comprehensive analysis of toxicity.

Table 2. Environmental impact categories in CML 2001.

Environmental impact category	**Units**
Abiotic depletion of elements (ADP-elements)	kg Sb eq./m^2
Abiotic depletion of fossil fuels (ADP-fossil)	MJ/m^2
Acidification potential (AP)	kg SO_2 eq./m^2
Eutrophication potential (EP)	kg PO_4^{3-} eq./m^2
Freshwater aquatic ecotoxicity pot. (FAETP)	kg DCB eq./m^2
Global warming potential (GWP)	kg CO_2 eq./m^2
Human toxicity potential (HTP)	kg DCB eq./m^2
Marine aquatic ecotoxicity pot. (MAETP)	kg DCB eq./m^2
Ozone layer depletion potential (ODP)	kg R11 eq./m^2
Photochemical ozone creation potential (POCP)	kg ethene eq./m^2
Terrestrial ecotoxicity potential (TETP)	kg DCB eq./m^2

2.3.2 Normalisation

We proposed a normalisation procedure according to the goal and scope of the LCA and based on the EPD developed by the EGGA (2016), which collected data from 66 companies in 14 countries in a sample representative of the European industry. Normalisation links the magnitude of the impact categories to reference values in order to frame the LCA results within a wider context and to compare the results in common dimensions (Margallo et al., 2014). Both the EGGA EPD (2016) and the present work used the CML 2001 environmental assessment method. Next **Fig. 2** shows the procedure to perform the normalisation of this work.

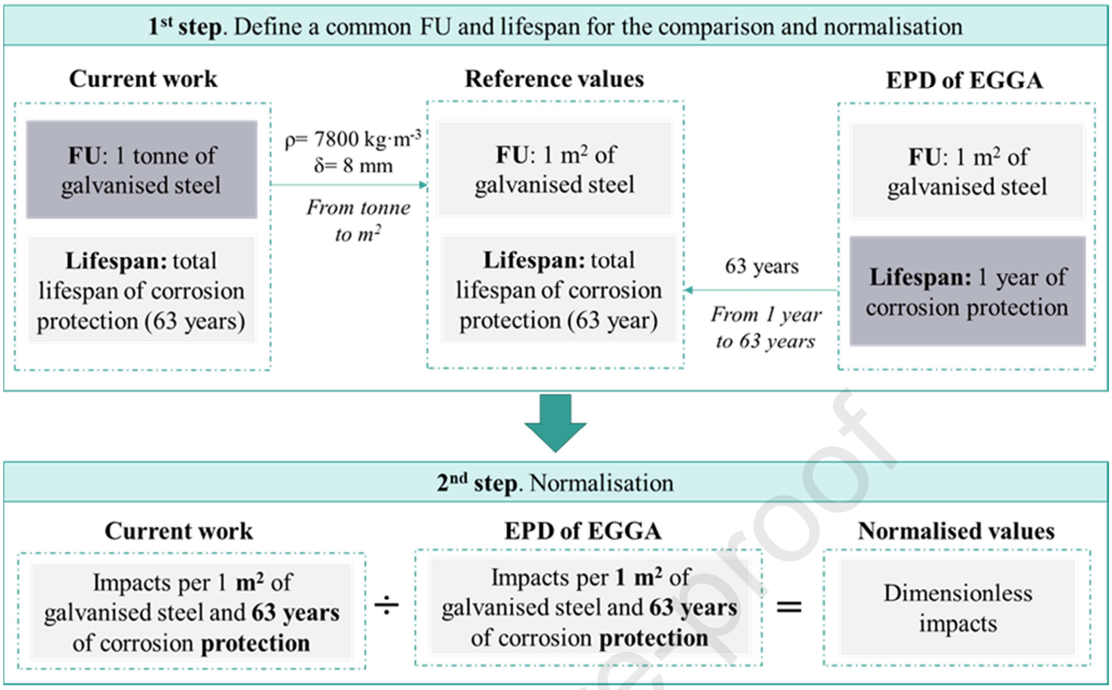

Fig. 2. Normalisation procedure.

The results of this work are referenced to one tonne of galvanised steel (**Table 4**). The first step to perform the normalisation is to adapt our results from tonnes to m^2 of galvanised steel. For this adjustment, we have used the density (7.8 tonne/cubic meter) and thickness (8 mm) of the galvanised steel material used by the EGGA in its EPD. The impact results of this work using 1 m^2 as FU and 63 years of corrosion protection are shown in Table S1.1 in the supplementary data. The second step is to obtain the threshold values using the environmental burdens reported by EGGA. In the EPD, the environmental impacts are shown for 1 year of corrosion protection and using 1 m^2 of galvanised steel as FU (Table S1.2). The threshold values for the normalisation are the EGGA results but considering 63 years as lifespan of the galvanised steel (**Table 3**), the same lifespan that is used in this work. Finally, the third step is to normalise the environmental burdens of this work (Table

S1.1) with the threshold values (**Table 3**), that resulted in the impact values shown in **Fig. 6**".

Table 3. Threshold values from the EGGA EPD (2016) for normalisation.

Environmental impact category	**Threshold value**	**Units**
Abiotic depletion of elements (ADP-elements)	$7.56\cdot 10^{-4}$	kg Sb eq./m^2
Abiotic depletion of fossil fuels (ADP-fossil)	1781	MJ/m^2
Acidification potential (AP)	0.34	kg SO_2 eq./m^2
Eutrophication potential (EP)	$7.43\cdot 10^{-3}$	kg PO_4^{3-} eq./m^2
Freshwater aquatic ecotoxicity pot. (FAETP)	-	kg DCB eq./m^2
Global warming potential (GWP)	163.8	kg CO_2 eq./m^2
Human toxicity potential (HTP)	-	kg DCB eq./m^2
Marine aquatic ecotoxicity pot. (MAETP)	-	kg DCB eq./m^2
Ozone layer depletion potential (ODP)	$9.10\cdot 10^{-7}$	kg R11 eq./m^2
Photochemical ozone creation potential (POCP)	$6.44\cdot 10^{-2}$	kg ethene eq./m^2
Terrestrial ecotoxicity potential (TETP)	-	kg DCB eq./m^2

3. RESULTS AND DISCUSSION

Here we present and discuss the main results of the LCA. Section 3.1 describes the environmental impacts of the HDG process and compares them against those of steel production. Secondly, the contribution of each stage in the HDG process to the impact categories is discussed in section 3.2, which also includes the comparison of the two Spanish HDG facilities. Finally, we normalise our results to those published by the EGGA to assess the situation of Spanish HDG plants within the framework of the European HDG sector.

3.1 Environmental performance of steel production and hot-dip galvanisation

Table 4 gives the total environmental impacts of the two Spanish HDG plants in 2017, as well as individual breakdowns of the impacts of steel production and hot-dip galvanising processes.

Table 4. Environmental impacts of steel production and hot-dip galvanisation in FU (1 tonne of galvanised steel) at HDG plants #1 and #2 in 2017.

		HDG plant #1			**HDG plant #2**		
Environmental impact category	**Units**	**Total**	**Steel sections**	**HDG #1**	**Total**	**Engineering steel**	**HDG #2**
ADP-elements	kg Sb eq./FU	$9\cdot 10^{-3}$	$-16\cdot 10^{-3}$	$25\cdot 10^{-3}$	$22\cdot 10^{-3}$	$-11.4\cdot 10^{-3}$	$33\cdot 10^{-3}$
ADP-fossil	MJ/FU	18,336	15,100	3,236	17,453	12,100	5,353
AP	kg SO_2 eq./FU	4.89	3.56	1.33	5.63	4.04	1.59
EP	kg PO_4^{3-} eq./FU	0.58	0.34	0.24	0.54	0.30	0.24
FAETP	kg DCB eq./FU	10.58	1.51	9.07	-11.93	-21.2	9.27
GWP	kg CO_2 eq./FU	1,719	1,440	279	1,475	1,030	445
HTP	kg DCB eq./FU	485	183	302	537	158	379
MAETP	kg DCB eq./FU	112,437	70,900	41,537	145,557	92,300	53,257
ODP	kg R11 eq./FU	$-6.21\cdot 10^{-7}$	$5.32\cdot 10^{-12}$	$-6.21\cdot 10^{-7}$	$-8.50\cdot 10^{-7}$	$1.19\cdot 10^{-11}$	$-8.50\cdot 10^{-7}$
POCP	kg ethene eq./FU	0.76	0.67	0.094	0.34	0.27	0.070
TETP	kg DCB eq./FU	8.97	2.56	6.41	12.30	3.79	8.51

To assess the differences between the two HDG plants in each impact category, **Fig. 3** shows the individual contribution (%) of steel production and the HDG process, considering that the total impact is the sum of both contributions.

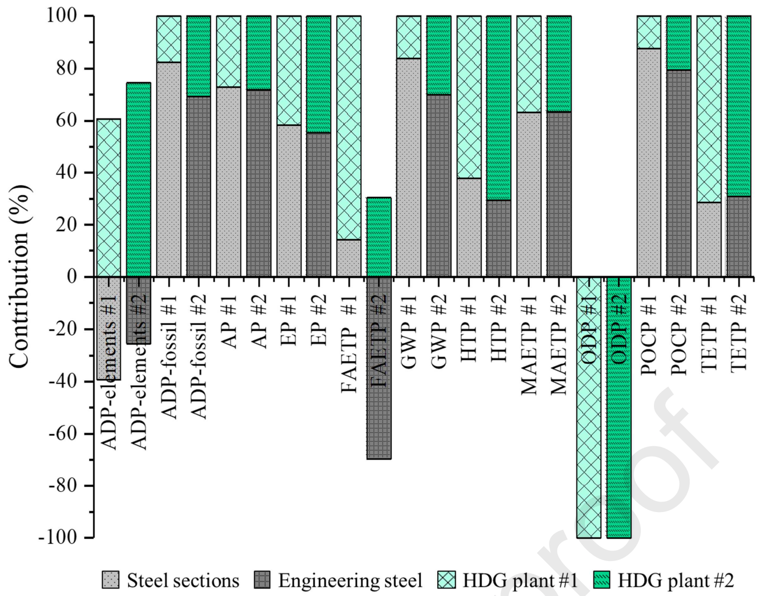

Fig. 3. Environmental impacts of steel production and the HDG process in 2017 for HDG plants #1 and #2.

The results in **Fig. 3** show that the impact categories ADP-fossil, AP, GWP, MAETP and POCP had a much greater contribution to steel production than galvanisation, which is more strongly influenced by ADP-elements, toxicity categories and ODP. Regarding steel production, the impact on ADP-fossil is due to the consumption of hard coal and natural gas, factors that make steel manufacturing one of the world's most polluting industries (Zhang et al., 2014). In addition to the use of carbon-based fuels, the large volume of steel produced worldwide and the high- energy use intensity make this industry as one of the main sources of CO_2 emissions (Rojas-Cardenas et al., 2017). As the EAF steelmaking process is less energy-intensive than the BOF process, the contribution of engineering steel to ADP-fossil at HDG plant #2 was lower than the impact of the steel sections used by HDG plant #1. The contribution to AP of emissions to air of nitrogen oxides (NOx) and sulphur dioxide (SO_2) in steel production was over 72% in both plants. Around 83% and

69% of the GWP at HDG plants #1 and #2, respectively, was caused by CO_2 emissions to air. The steel production's contribution to MAETP was over 63% at both plants because of hydrogen fluoride (HF) air emissions. Finally, about 88% and 79% of POCP at HDG plants #1 and #2 resulted from CO_2, SO_2 and NOx air emissions from the steelmaking processes. The contributions of steel production and galvanisation were similar for the impact category EP. About 58% and 55% of the impact in HDG plants #1 and #2 was associated with gaseous emissions of inorganic compounds, mainly NOx, from steel production.

On the other hand, the categories in which galvanisation contributed more than steel production were ADP-elements, FAETP (only in HDG plant #1), HTP and TETP. So, it seems that galvanisation has a greater influence over toxicity impact categories. The contribution of galvanisation to ADP-elements was related to silver's impact on zinc production, as explained below. Primary zinc production involves mining, ore beneficiation and zinc metal refining, which can be carried out by electrometallurgical or pyrometallurgical smelting (Van Genderen et al., 2016). Of these processes, the electrometallurgical method is used in over 95% of the world's zinc refineries. Zinc is never found in isolation in its mineral ores, but rather it is commonly found with other elements (Farjana et al., 2019). Zinc ore processing produces iron, lead, silver, copper, cadmium and indium concentrates in addition to the zinc concentrate (Van Genderen et al., 2016). In **Fig. 3**, the impact of HDG in terms of ADP-elements derives from silver. Silver is a by-product of zinc production, but this assessment is not considered in the database. Galvanisation contributed more than 85% of FAETP at HDG plant #1 due to the emission of heavy metals to freshwater derived from zinc production. With respect to HTP, galvanisation accounted for 62% of the impact due to the emission of arsenic (+V), cadmium and copper to air from zinc production at HDG plants #1 and #2. In HDG plant #2, HTP was also influenced by organic emissions, mainly polycyclic aromatic hydrocarbons (PAH). On another note, almost 70% of TETP at both HDG plants was caused by the emission of lead and other heavy metals to air during zinc production.

Finally, **Table 4** and **Fig. 3** include impact categories with a negative value for ADP-elements, FAETP (only at HDG plant #2) and ODP. These data should be treated with caution, as the negative impacts should not be interpreted such that an increase in product consumption would lead to any reversal of environmental burden elsewhere (Van Genderen et al., 2016). Steel production had a negative impact on ADP-elements for both plants and on FAETP at HDG plant #2. ADP-elements was negative due to the system expansion to include co-products of the steelmaking processes. Co-products are defined as any of two or more products from the same unit process or system (European Comission et al., 2010). In this case, we considered lead to be a co-product recovered from the production of primary zinc. The material credits associated with this consideration were $-2.06\cdot10^{-3}$ and $-3.56\cdot10^{-3}$ kg of lead per kilogram of steel sections and engineering steel, respectively. At HDG plant #2, the environmental impact of steel engineering resulted in a negative value in the FAETP category. This contribution is negative because steel engineering production is based on EAF steelmaking which uses secondary steel as input (Yellishetty et al., 2011). Scrap and metal dust contain heavy metals such as nickel and copper that are reprocessed when producing steel. Specifically, the emission credits associated with engineering steel production were $-4.65\cdot10^{-6}$ kg nickel/kg steel and $-6.58\cdot10^{-6}$ kg copper/kg steel, which influenced the impact category FAETP. Finally, ODP impacts were negative at both HDG plants due to the galvanisation process. Primary zinc production led to negative organic emissions to air because the collection and incineration of refrigerant gases such as trichlorofluoromethane (R11), dichlorotetrafluoroethane (R114) and dichlorodifluoromethane (R12) was taken into account, which entails energy credits (Van Genderen et al., 2016).

3.2 Stages and impacts of hot-dip galvanisation

This section analyses the impacts of each stage of the galvanisation process. The additional consumptions stage also includes the water and electricity inputs. **Fig. 4** shows the results for HDG plants #1 and #2 as the contribution (%) of each stage to the total impact value, whereas the absolute values are collected in Tables S3.1 and S3.2 (supplementary data).

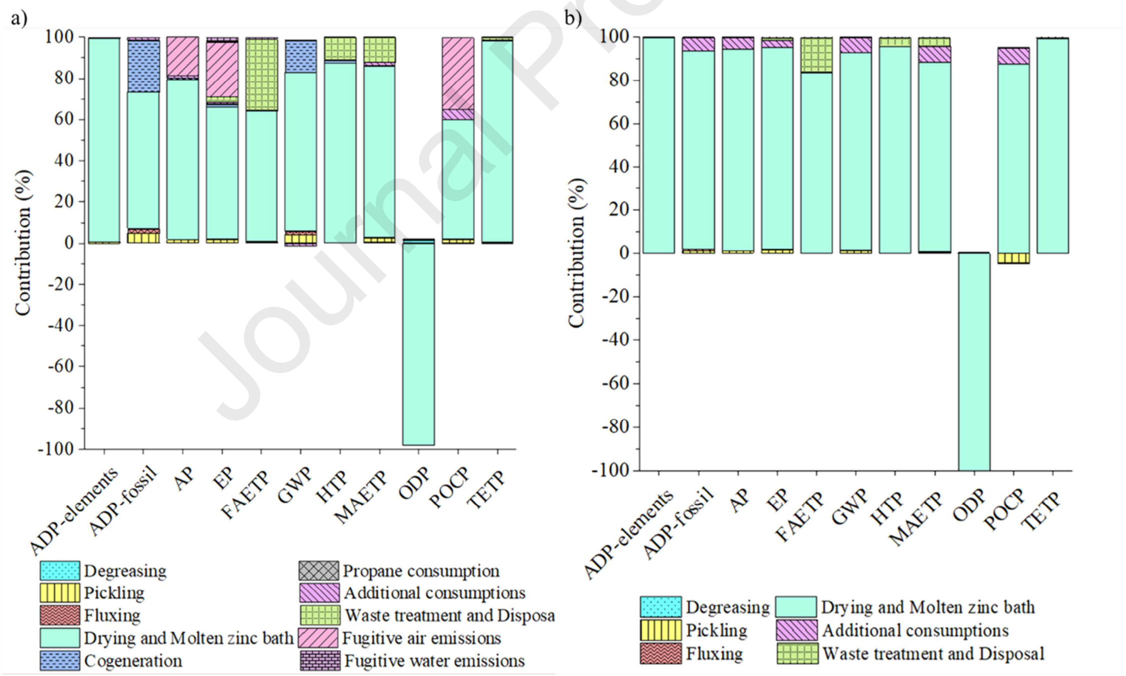

Fig. 4. Environmental impact of a) HDG plant #1 and b) HDG plant #2 by stages, in 2017.

Fig. 4 shows that the drying and molten zinc bath stage contributed the most to all the impact categories at HDG plants #1 and #2. These results are due to the significant impact of primary zinc production, which is the main material input in molten zinc baths. The rest of the HDG stages had less influence on the impact categories being studied. Section 3.1 explained the consequences of primary zinc production on ADP-elements, FAETP (only at HDG plant #1), HTP and TETP. Next, we explain the influence of primary zinc production on the rest of impact categories. ADP-fossil was due to the hard coal and natural gas consumption required when processing ores to extract and refine primary zinc (Norgate et al., 2007), although these consumptions were lower for HDG plant #1 than HDG plant #2 (66% compared to 92%). The contributions of NOx and SO_2 emissions during zinc production were 77% and 64% with respect to AP and EP for HDG plant #1 and 93% for both impact categories at HDG plant #2. The FAETP impact at both plants was influenced by heavy metal emissions to freshwater of 63% and 83% at HDG plants #1 and #2 respectively. GWP is caused by CO_2 emissions in primary zinc production, with contributions of 79% and 91% at HDG plants #1 and #2 respectively. MAETP is explained by air emissions of heavy metals (copper and vanadium) and inorganic air emissions in the form of hydrogen fluoride, wherein the drying and molten zinc bath stage contributed 83% and 88% for HDG plants #1 and #2. This indicates that biologically essential metals, but which are toxic at high concentrations (Norgate et al., 2007), affect the toxicity impact categories. Regarding ODP, the results were 100% negative because of the energy credits associated with the incineration of the refrigerants (R11, R114 and R12) used during zinc production, as explained in section 3.1. Finally, almost 58% and 97% of POCP was related to zinc production in the form of NOx and SO_2 emissions at HDG plants #1 and #2 respectively.

However, **Fig. 4a** shows that some impact categories for HDG plant #1 were also influenced by other stages of the galvanisation process other than the molten zinc bath. Specifically, 25% of ADP-fossil and 16% of GWP were assigned to the cogeneration unit that involves natural gas consumption and the consequent CO_2 emissions. What is more, fugitive air emissions contribute 35% to POCP (NOx and CO), 19% to AP and 26% to EP (NOx and CO_2). Moreover, waste treatment and disposal have a 35% influence on the impact category FAETP and around 11–12% contribution to HTP and MAETP. Waste treatment involves heavy metal emissions to freshwater which increase the stage's influence on the toxicity impact categories HTP and MAETP. On the other hand, **Fig. 4b** shows that waste treatment and disposal also influenced some categories for HDG plant #2. In particular, this stage had a 16% contribution to FAETP and 4% to HTP and MAETP due to the freshwater emission of thallium. Additional consumptions involved a small contribution, although they were significant in the categories ADP-fossil, AP, EP, GWP, MAETP and POCP because of the associated electricity consumption. Furthermore, the acid degreasing and pickling stages produced a negative POCP at HDG plant #2 because of nitrogen monoxide (NO) emissions derived from the use of trucks in transport. NO has an odd number of electrons, which means NO is a very reactive molecule (Bange, 2008). In the troposphere, NO is rapidly oxidised to either nitrous acid (HNO_2) or nitrogen dioxide (NO_2). In the latter case, NO reacts with photochemical ozone so it has a negative contribution to POCP.

From the above analysis it seems clear that mitigating the impact of HDG would require measures designed to reduce raw material consumption (zinc) and waste generation. Almost 94% of the impacts of waste treatment come from the spent pickling acids. In this regard, a few research initiatives are promoting innovative work looking at zinc recovery from spent pickling acids for reuse in the HDG process (Arguillarena et al., 2019; Laso et al., 2015; LIFE2ACID, 2020).

Fig. 5 summarises the comparison of the environmental performance of the two Spanish HDG plants in 2017, excluding their impacts associated with steel production.

In general terms, the environmental impacts of the HDG process was greater at plant #2, apart from POCP, for which fugitive air emissions caused an increase in this impact category at HDG plant #1. The differences between the two plants can be attributed to the use of a greater amount of primary zinc per FU at HDG plant #2. **Table 1** shows that HDG plant #2 consumed 79 kg of zinc per tonne of galvanised steel, while in HDG plant #2, zinc usage was reduced to 59 kg of zinc per tonne of galvanised steel. Similarly, the better performance of HDG plant #1 in ADP-fossil is associated with the optimal efficiency of the cogeneration unit in the combustion of natural gas to produce thermal energy and electricity.

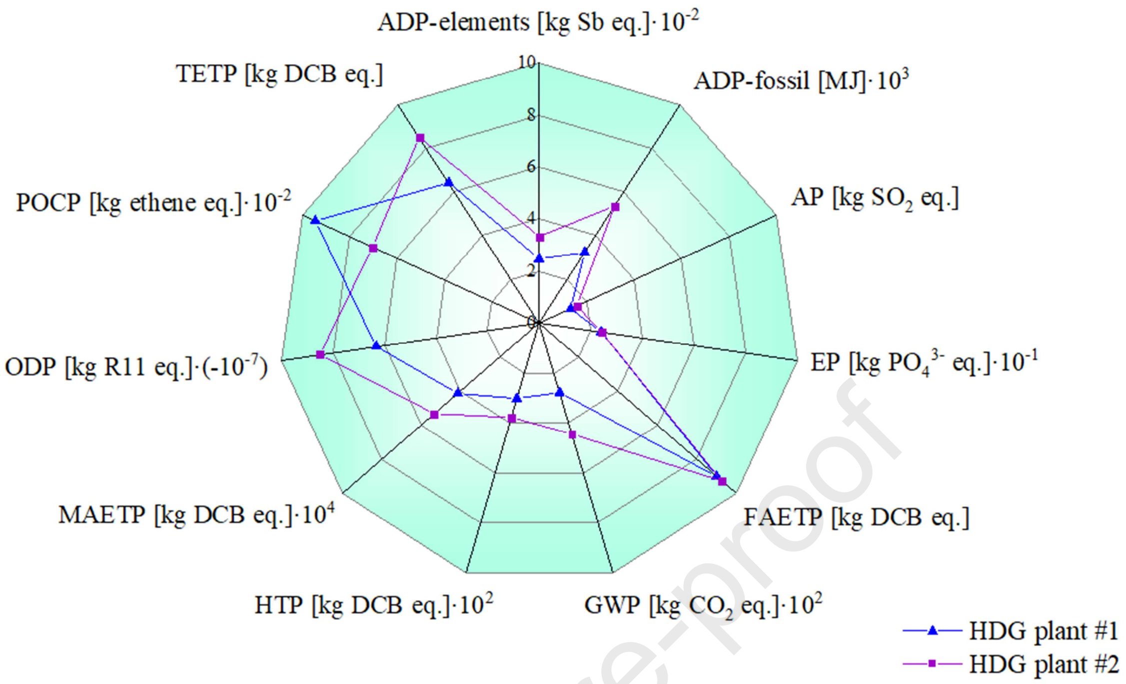

Fig. 5. Environmental impacts of plants HDG #1 and #2 in 2017.

3.3 The Spanish hot-dip galvanisation process in a European setting

This section compares the environmental impacts of the two Spanish HDG case studies with the results reported by the EGGA (EGGA, 2016), which do not include the toxicity impact categories HTP, MAETP, FAETP and TETP. To make this comparison we first normalised our data to place the Spanish galvanisation industry within the European framework. The normalisation procedure was explained in detail in section 2.3.2. **Fig. 6** shows the final normalised results for both HDG plants and the EGGA reference values.

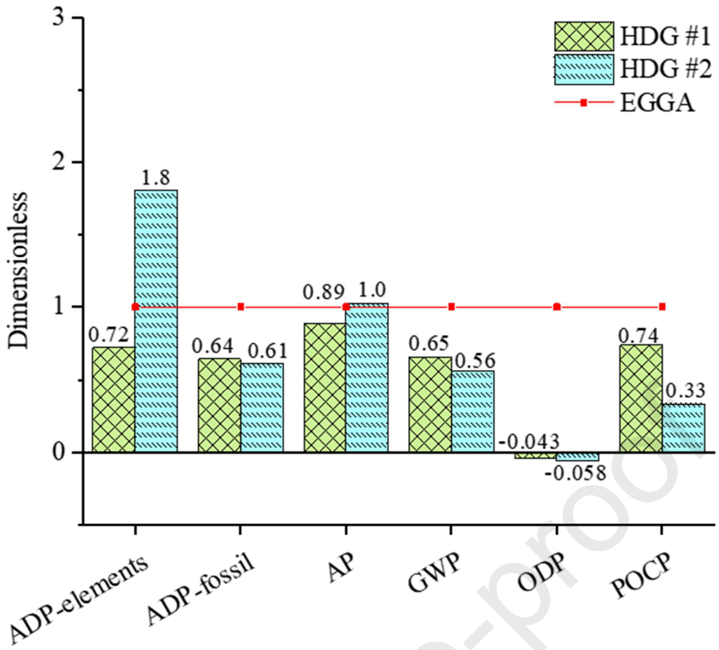

Fig. 6. Normalisation results using data obtained by the EGGA (2016) as reference values.

Despite implementing the same LCIA method in both the EGGA report and the present study there are some differences that should be addressed when interpreting the results. The EGGA report used web-based questionnaires to collect data from 66 companies in 14 countries. The sample considered the production of a range of light, medium and heavy products with a small contribution from centrifugal galvanisation (EGGA, 2016). The EGGA analysis also examined the manufacturing, distribution, waste transport and product disposal stages. The distribution had very little effect on resource usage and water consumption, and, therefore, on the environmental impact, which is why we did not include this stage in our study. With regard to the LCA method, it is worth remembering that the EGGA published their work in 2016 and, therefore, some of the characterisation factors could be out of date. Moreover, the two studies use of different databases and unit processes is another source of uncertainty. The results of the normalisation show that both

Spanish HDG plants were below the EGGA values in most of the impact categories. Exceptions were found in ADP-elements at HDG plant #2, which almost doubles the European value. ADP-elements results completely depend on primary zinc production, and while HDG plant #1 used primary zinc more efficiently than the European values, plant #2 had a higher consumption of raw materials. The EP category was initially included in the normalisation, but the impact of HDG plants #1 and #2 were well above the EGGA values. This difference can be attributed to updated data in our study for NOx emission characterisation factors, which are the main driver of the EP category, so we excluded this impact category from the assessment. The impact category ODP represents one of the main differences with the EGGA study. Unlike the EGGA study, our ODP results were negative because we included the energy credits associated with waste incineration, as explained in sections 3.1 and 3.2. Steel production is a decisive stage in the rest of the impact categories. This is particularly evident in the case of ADP-fossil, GWP and POCP. The use of different types of steel has a significant influence on the results. In this sense, the EGGA report employed steel plate to represent the European average instead of steel sections, which are representative of the steel components galvanised at HDG plant #1, and engineering steel, which is mostly used in the components galvanised by HDG plant #2. We assessed the environmental impacts of these types of steel using the World Steel database. The results for steel plate, steel sections and engineering steel per tonne of galvanised steel are 26,500, 15,100 and 12,100 MJ for ADP-fossil; 2,600, 1,440 and 1,030 kg CO_2 eq. for GWP; and 0.808, 0.668 and 0.270 kg ethene eq. for POCP. Therefore, the use of steel sections and engineering steel reduced the impact by 43–54% in ADP-fossil, by around 46–60% in GWP and 24–69% in POCP. This confirms that steel plate has a greater impact than other types of steel and, hence, potentially explains the differences in the normalised results.

Generally, the environmental performance of the Spanish galvanisation plants was better than the EGGA values. However, the environmental results are highly dependent on the type of steel being galvanised and should be viewed with caution.

4. CONCLUSIONS

In the current study, LCA methodology was used to assess the environmental performance and to identify hotspots of the hot-dip galvanising process from cradle-to-gate. Two representative Spanish HDG plants were selected to identify the contribution of each stage of the HDG process including toxicity impact categories for the first time. Besides, the external normalisation proposed allows placing the Spanish HDG plants at the European level in a simple way. This type of normalisation can be used in the decision-making process to improve the sustainability of HDG.

The most relevant environmental impacts of the HDG process at both plants were due to steel and primary zinc production and energy consumption. The impact of steel varies from type to type and this is reflected in the results since each of the HDG plants galvanised special components using different types of steel ("steel sections" and "engineering steel"). On the other hand, primary zinc production was the main contributor to the HDG process' environmental impact because it is a very resource- and energy-intensive process. The consumption of primary zinc and natural gas in the drying and molten zinc bath makes these stages the ones with the greatest impact. In this context, the consideration of by-products and/or co-products in the steel and zinc production can affect the environmental results providing uncertainty. In general, both HDG plants were in a better environmental performance compared to the European values but the selection of a representative type of steel has a significant influence on ADP-fossil, GWP and POCP.

This study lays the foundations for improving the resource efficiency and productivity of galvanisation plants. Mitigating the impact of HDG would require designing measures to reduce raw material consumption (zinc) and waste generation. The use of primary zinc should be as efficient as possible. Secondary zinc usage is a potential option because its production requires less energy and resources, while also guaranteeing the quality of the final product. Furthermore, the sustainable management of the baths used in the different stages of the HDG process could help to extend their lifespan and therefore reduce the rate of resource consumption. All of these factors bring us closer to the transition to more sustainable HDG processes. Future developments of this work would extend the system boundaries to include the recovery of zinc from ash and dross. In addition, the LCA of the zinc recovery from spent pickling acids by membrane based solvent extraction and electrowinning will be performed as the project LIFE2ACID proposes.

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Highlights

- The hot-dip galvanisation (HDG) sector in Spain was assessed via LCA.
- Two HDG plants with different production properties provided detailed data inventories
- The highest impacts were found in the production of steel and primary zinc.
- Using secondary zinc and extending pickling bath´s lifespan would improve HDG
- The impacts of the Spanish HDG plants were generally below the European average

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: