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Coupling phytoremediation of Pb-contaminated soil and biomass energy production: A comparative Life Cycle Assessment

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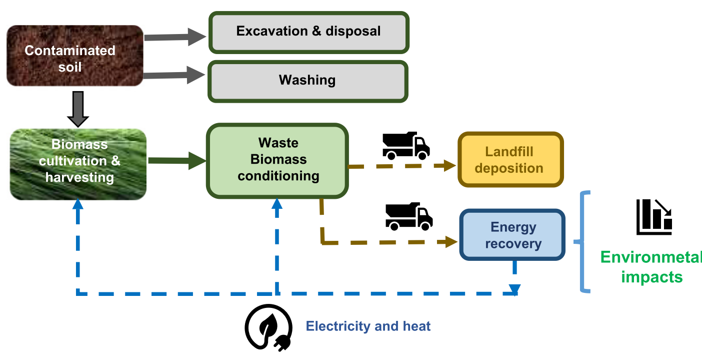
HIGHLIGHTS

- LCA methodology applied to soil remediation processes.
- Phytoremediation with *Festuca arundinacea* is efficient for Pb-contaminated soil.
- Energy recovery reduces impacts regards other biomass disposal treatments.
- Phytoremediation + energy recovery shows the best environmental footprint.

GRAPHICAL ABSTRACT



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ABSTRACT

Phytoremediation is an in-situ remediation technology based on the ability of plants to fix pollutants from the soil. In this sense, plants such as *Festuca arundinacea* are a promising for heavy metal removal in contaminated soils. The present work studies phytoremediation for Pb removal from a contaminated soil located in Spain using *F. arundinacea* by applying the Life Cycle Assessment (LCA) approach. Two different options for biomass management were assessed: direct disposal in a security landfill (case 1A) and energy recovery (case 1B). For the latter option, cogeneration was simulated using SuperPro Designer 9.5. In addition, traditional treatments such as soil washing (case 2) and excavation + landfill (case 3) were evaluated in terms of environmental impacts by LCA. The former was simulated using SuperPro Designer 9.5, whereas data from literature were used for the latter to perform the LCA. Results showed that biomass disposal in a landfill was the most important contributor to the overall impact in case 1A. In contrast, biomass conditioning and cogeneration were the main steps responsible for environmental impacts in case 1B. Comparing cases 1A and 1B, the energy recovery from biomass was superior to direct landfill disposal, reducing the environmental impacts in most of the studied categories. Regarding the rest of the treatments, chemical production and soil disposal presented the most critical environmental burdens in cases 2 and 3, respectively. Finally, the comparison between the studied cases revealed that phytoextraction + energy recovery was the most environmentally friendly option for the studied conditions, reducing impacts by 30–100%.

1. Introduction

Nowadays, environmental contamination by heavy metals is a global priority concern. These pollutants are non-biodegradable, carcinogenic and mutagenic compounds and consequently, they cause harmful effects on the environment and human health (Gluhar et al., 2020). In this sense,

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contaminated soils with heavy metals induce plants to accumulate those compounds compromising human health through food chains (Alaboudi et al., 2018; Wang et al., 2020). For this reason, governments have established strict regulations on the content of heavy metals in soils (Gluhar et al., 2020; Khalid et al., 2017).

Concerning heavy metals, lead is one of the most polluting metals distributed in soil due to mining and smelting activities, automobile emissions related to gasoline combustion, battery disposal, chemical production and factory emissions (Fan et al., 2020; Hasegawa et al., 2019; Lei et al., 2020; Liu et al., 2018; Wang et al., 2020). This metal is considered one of the most harmful to the environment as it can negatively affect plant growth and metabolic activities. Regarding human health, lead can cause damage to organs and the nervous system (Barbafieri et al., 2017).

A wide variety of treatments to remove heavy metals are available for soil remediation, although selecting the most suitable alternative is difficult due to technical limitations (Vocciante et al., 2019). The reported technologies to remove these contaminants could be classified into three groups: physical (soil replacement, soil isolation, vitrification and electrokinetic, etc.), chemical (immobilization, stabilization/solidification, soil washing, etc.), and biological (phytoextraction, phytostabilization, biosorption, etc.) (Alaboudi et al., 2018). In turn, these treatments can be classified concerning their application place: in-situ in the contaminated area or ex-situ (if the soil is excavated and further treated in another site) (Gnanasundar and Akshai Raj, 2020; Hasegawa et al., 2019; Liu et al., 2018). Currently, the most usual strategies for heavy metal removal from soils are physical (excavation and landfill disposal) and chemical (soil washing) (Gluhar et al., 2020; Gong et al., 2018; Kim et al., 2013). However, they present important drawbacks, mainly related to their cost and/or their social and environmental impacts (Alaboudi et al., 2018; Khalid et al., 2017; Vocciante et al., 2019).

Phytoremediation is an in-situ remediation technology based on the ability of plants to fix pollutants from the soil under the concept of using nature to cleanse nature ((Erakhrumen, 2007). This approach can replace clean-up technologies that are often laborious and costly. Phytoremediation involves phytoextraction, phytostabilization and phytovolatilization processes. Among them, phytoextraction is based on the ability of plants to remove toxic metals from soil or water through their uptake into the harvestable parts of the plant (Suman et al., 2018). These treatments effectively remediate soils containing and, in addition, they allow recycling the clean soil (Alaboudi et al., 2018). Furthermore, the obtained biomass can be subsequently used to different ends, mainly related to energy recovery. Nevertheless, phytoextraction of toxic metals presents limitations regarding the types of plants that can be used, being the preferred option for hyper-accumulator plants capable of fixing high concentrations of heavy metals (Barbafieri et al., 2017). In this sense, different species of plants (*Amaranthus* spp., *Medicago sativa*, *Sorghum bicolor*, *Brassica nigra*, *Pisum sativum*, *Brassica juncea*, *Salix viminalis*, *Helianthus annuus*, *Sinapis alba*, *Zea mays*, *Festuca arundinacea*, etc.) can be used to remove a wide range of heavy metals (As, Cd, Ni, Pb, etc.), as reported in the literature (Al-Jobori and Kadhim, 2019; Govarthanan et al., 2018; Mahar et al., 2016; Razmi et al., 2021; Yashim et al., 2015; Zehra et al., 2020; Zhou et al., 2020). These plants can accumulate high contents of heavy metals in their tissues despite their low cultivation yield.

In this context, *F. arundinacea* seems to be a promising option for heavy metal removal in soils, as it is reported in recent studies (Steliga and Kluk, 2020; Wasilkowski et al., 2019). *F. arundinacea* is widely grown as turfgrass and forage, in temperate regions. It has strong roots and can grow in many soil types (Feng et al., 2018). These species present fast growth with high biomass productivity that can accumulate metals in their tissues. Besides, during the state of vegetation, this species shows high tolerance to heavy metals like Pb, Cd, Cu, Zn and Ni (Hu et al., 2015; Lou et al., 2017; Lu et al., 2014; Steliga and Kluk, 2020). Concerning lead, *F. arundinacea* shows a higher translocation factor (TF) between shoots and roots (ability to transfer the metal from roots to shoots) than for other metals (Steliga and Kluk, 2020). Furthermore, it has been reported the possibility to recover energy (electricity and heat) from *F. arundinacea* using different technologies such as anaerobic digestion (Feng et al., 2018; Molinuevo-Salces et al., 2014), pyrolysis (Fahmi et al., 2007) or cogeneration (Cristaldi et al., 2017). These features make *F. arundinacea* a promising alternative for phytoextraction of Pb from contaminated soils.

As above mentioned, one of the most controversial aspects of soil remediation processes is their environmental sustainability. Life Cycle Assessment (LCA) is a methodological approach to quantify the environmental impacts of products and processes (Espada et al., 2021). This approach is suitable to determine the environmental feasibility of different technologies for soil remediation (Vocciante et al., 2019, 2021). In this sense, many LCA studies on soil remediation processes have been reported in the literature (Chen et al., 2020; Harris et al., 2016; Visentin et al., 2019). The application of LCA to phytoremediation of soils contaminated by heavy metals has been reported previously (Vigil et al., 2015; Witters et al., 2012), but it is scarce. Recently, Vocciante et al. (2019) have reported a study on the carbon footprint, including both the phytoextraction of heavy metals with different plants (*Brassica juncea*, *Lupinus albus* and *Helianthus annuus*) as well as the further use of biomass to obtain energy (Vocciante et al., 2019). Furthermore, these authors have also reported the comparison of these phytoextraction treatments with other methods, but only in terms of CO_2 emissions (Vocciante et al., 2021). The same approach was reported by Todde et al. (2022), which applied LCA to phytoremediation of polluted soils using *Cannabis sativa* L. and further energy recovery from biomass, including results of Cumulative Energy Demand and CO_2 footprint (Todde et al., 2022). In this context, it is necessary to enhance the knowledge on the environmental footprint of phytoremediation technologies for soils contaminated by heavy metals in order to identify main environmental burdens as well as establish potential improvements of this technology compared to other remediation treatments.

In the present work, phytoremediation of a highly Pb-contaminated soil from an old abandoned mine located in Spain using *F. arundinacea* was studied by applying the LCA approach. For that purpose, data on cultivation and Pb uptake of *F. arundinacea* were calculated from the literature, and different final uses of biomass were studied: security landfill disposal and energy recovery (this option was simulated using SuperPro Designer 9.5). The environmental impacts of both scenarios were quantified by using Gabi 10 software. On the other hand, the LCA of two traditional treatments (soil washing and excavation) was performed. For this purpose, the former treatment was simulated using SuperPro Designer 9.5, whereas literature data were adapted for the latter to quantify their environmental impacts. Finally, the comparison of LCA results between phytoremediation-based scenarios and traditional treatments was carried out.

2. Materials and methods

2.1. Soil characterization

The contaminated soil considered in this work is a highly Pb-contaminated soil close to an abandoned mine in a Mediterranean area in Spain. The studied soil corresponds to an old mining area located in the region of Barberà basin, in the south of Catalonia, Spain. It is located in the SW area of the Catalonian mountain range which extends parallel to the Mediterranean coast. The pluviometric information of this area (last 10 years was taken from Servei Meteorològic de Catalunya and it is shown in Table S1 of the Supplementary material (Servei Meteorològic de Catalunya, 2022). It presents an average Pb concentration of 180 mg kg^{-1} (dry soil basis) at an average depth of 20 cm (Durán Cuevas, 2010). The objective of the study is to reduce Pb concentration to 60 mg kg^{-1} of soil in order to fit the regulations of the Local Government for urban and/or agricultural use of the soil (Spanish Government, 2009). The average texture of the soil was taken from data on this soil available in the literature (Durán Cuevas, 2010): 61% of sand particles (2–0.05 mm), 28% of silt particles (0.05–0.002 mm) and 11% of clay particles (<0.002 mm). According to this information, the soil can be considered as sandy loam type (United States Department of Agriculture, 2022). The pH of the soil was 5.7 (Durán Cuevas, 2010).

2.2. Goal and scope of the LCA study

This study aims to determine the environmental performance of phytoextraction with *F. arundinacea* for the remediation of the above soil by applying LCA methodology. For that purpose, three treatments were assessed: phytoextraction (study case 1), washing soil (study case 2), and excavation and landfill deposition (study case 3). Fig. 1 shows an overview of the case studies, including the involved steps. All the inputs and outputs of the processes were considered within the system boundaries, whereas capital goods are excluded of the study. On the other hand, the labor machinery was considered as fuel consumption for cultivation operations (plowing, harrowing, sowing and harvesting) in the phytoremediation case. The fuel consumption was calculated according to the specific characteristics of the machinery, using spreadsheets reported by the Spanish Government for agricultural labor (Spanish Government, 2020).

The functional unit considered for all the studied treatments was the decontamination of 1 ha of the considered soil (at a depth of 20 cm) up to the maximum Pb concentration allowed by the government regulations. The amount of soil equivalent to the functional unit was calculated from its apparent density (1500 kg m^{-3} for this type of soil (Antúnez et al., 2015), resulting in a value of 3000 tons.

2.3. Life Cycle Inventory Analysis (LCIA)

Inventory data of the three case studies (involving all the steps) were calculated and adapted from the literature and/or simulated using SuperPro Designer 9.5 (Intelligen Inc. Scotch Plains, NJ, USA) as described below for each case. Primary processes of energy and material production were taken from Gabi Professional 2021 database (Sphera Solutions GmbH. Leinfelden-Echterdingen, Germany). For electricity production,

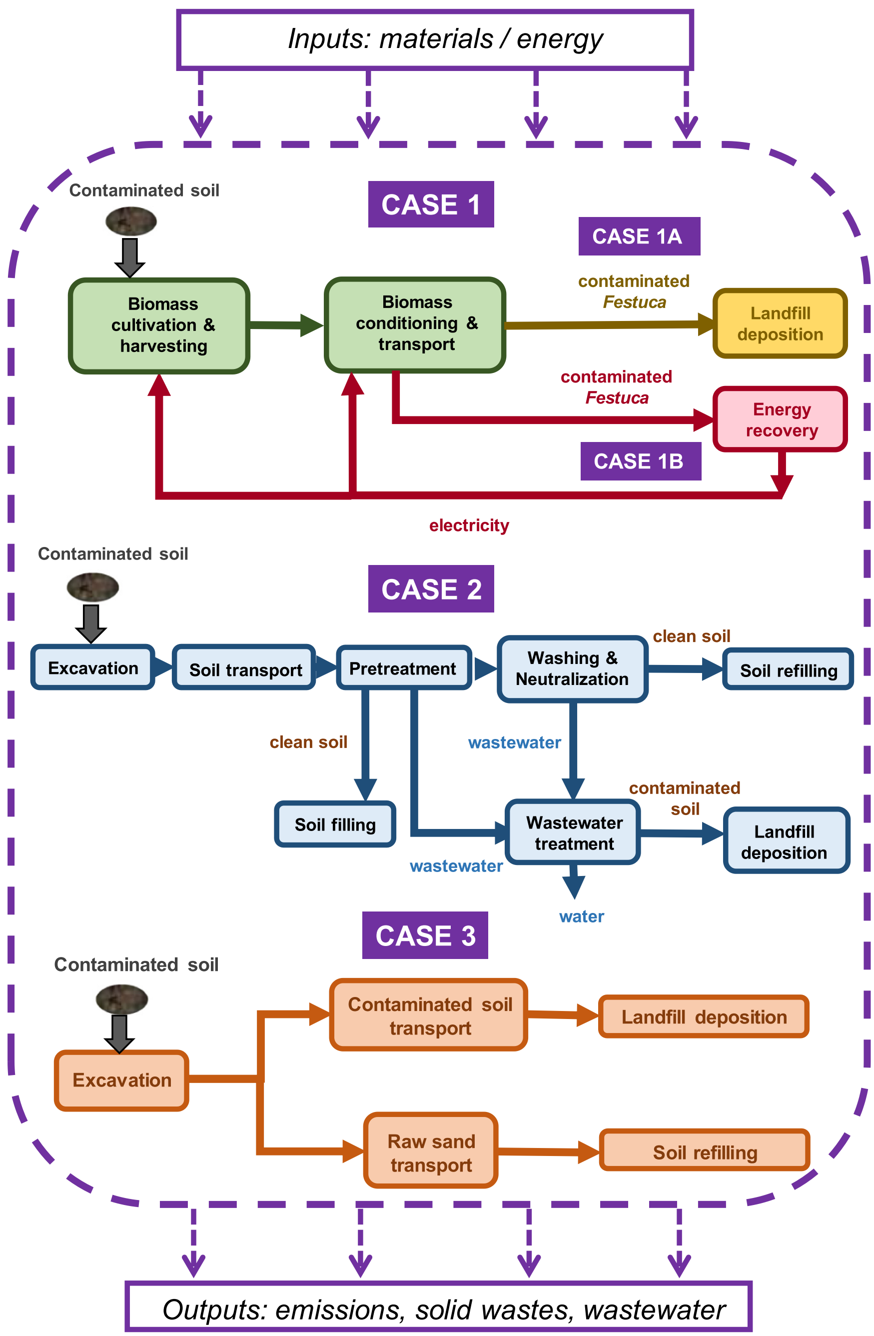

Fig. 1. LCA limits of the considered case studies.

the Spanish electric mix updated to the current year was used. Transportation vehicles were adapted from inventory data available in Gabi Professional 2021 database.

2.3.1. Case study 1: phytoremediation with Festuca arundinacea

The main steps in the phytoextraction of Pb-contaminated soils using *F. arundinacea* (Fig. 1) are cultivation and harvesting, conditioning and transport and use of the grown biomass. In this case, two possibilities for biomass use were analyzed: landfill deposition or energy recovery by cogeneration in a combined power and heat (CPH) system (cases 1A and 1B, respectively). In the latter scenario, the cogeneration process was simulated using SuperPro Designer 9.5. A detailed scheme of the flowsheet diagram is shown in the supplementary material (Fig. S1), and the detailed inventory data of both scenarios are summarized in Table 1. The detailed steps involved in case 1 are the following:

i. *Biomass cultivation & harvesting*: Initially, it is necessary to improve soil conditions by controlling weeds and increasing the soil porosity by plowing. The amount of fuel required for the agricultural machinery was calculated according to their specific characteristics, using spreadsheets reported by the Spanish Government for agricultural labor (Spanish Government, 2020). These values are summarized in Table 1.

As above described, the cultivation of *F. arundinacea* is enhanced under Mediterranean conditions. In this work, a seed rate of *F. arundinacea* of 30 kg ha^{-1} was assumed according to the literature (Delgado, 1984). The total amount of seeds is shown in Table 1. The fuel consumption of the seed drill was calculated according to the specific characteristics of the machinery, using spreadsheets reported by Spanish Government for agricultural labour (Spanish Government, 2020). Regarding to land fertilization requirements (twice per year), the recommendation for soil fertilization NPK 90:100:50 (kg ha^{-1}) was taken (Bazzigalupi and Bertín, 2014; Delgado, 1984), and the values are shown in Table 1. Finally, a value of 1.75 kg CO_2 ha^{-1} was assumed for CO_2 fixation by *F. arundinacea* (Flores, 2017).

F. arundinacea accumulates more amount of lead in roots than in shoots, as shown by the bioconcentration factor (BCF) values: 3.73 (roots) and 1.84 (shoots), respectively (Steliga and Kluk, 2020). In the present work, *F. arundinacea* was cultivated in spring, and the biomass was harvested in two cuts during the vegetative period, according to the recommendation reported in the literature (Formoso, 2010; Ramírez Fonseca, 2011; Steliga and Kluk, 2020; Tilvikiene et al., 2016). At the end of the vegetative period, the biomass is withdrawn along with the roots. In all cases, biomass harvesting was carried out using a combine forage harvester with trailer. Irrigation requirements for *F. arundinacea* are not taken into account because the soil is located in a region with an average rainfall from March to November of around 50 mm, similar to other regions in which *F. arundinacea* is cultivated in the same way (Delgado, 1984; San Miguel, 2008; Tilvikiene et al., 2016). Rainfall data are summarized in Table S1 of Supplementary material.

Having in mind the total amount of biomass (24.120 kg ha^{-1} $year^{-1}$) along with the experimental root/shoot ratio of 0.9 (in agreement with previous data reported (Crush et al., 2012), biomass from roots and shoots was calculated. Then, Pb extracted by roots and shoots were calculated using BCF as commented above). In these conditions, the amount of Pb extracted per year was 11.59 kg ha^{-1}, resulting in a total duration of 31 years to remove the targeted amount of Pb (358 kg ha^{-1}) to fulfill local government regulations. The biomass required to remove this Pb amount is summarized in Table 1.

ii. *Biomass conditioning and transport:* To remove the soil contained in the biomass, a vibration screening was added to calculate its energy consumption by simulation using SuperPro Designer 9.5 according to the flowsheet shown in Fig. S1 (Supplementary material). The inventory data for transport were adapted from Gabi Professional 2021 database assuming 30 km between the contaminated area and the treatment plant for biomass disposal.

iii. *Biomass use:* One of the key aspects concerning the sustainability of phytoextraction processes is the further use of the grown biomass (Vocciante et al., 2021). In this work, two possibilities are raised:

A) *Soil disposal*: In this scenario, the harvested biomass is disposed as hazardous material in an existing landfill 30 km away from the contaminated area. The landfill was supposed as an underground deposit adapted from Gabi Professional 2021 database.

B) *Energy recovery*: In this case, the harvested biomass is used to produce energy through a combined power and heating system (CPH). In order to calculate the inventory data of this step, the process was simulated using SuperPro Designer 9.5 (Fig. S1, supplementary material). As mentioned above, the biomass is transported from the contaminated area until the CPH plant (the same distance as to landfill was supposed). Then, the moisture of the biomass is reduced in an air oven before passing to a boiler in which the biomass is stoichiometrically burnt in the presence of air, obtaining steam, off-gas and ashes (rich in Pb). For this purpose, the molecular formula of the biomass was calculated from elementary analysis of *F. arundinacea* reported in literature (Fahmi et al., 2007). The steam obtained is subsequently expanded in a multistage turbine, producing electricity and heat with an efficiency of 35% and 50%, respectively, according to the literature (Scano et al., 2014). The ashes obtained in the combustion process are transported and disposed in a security landfill for hazardous wastes (underground deposit). Table 1 summarizes the obtained results for the duration of the treatment.

Table 1
Inventory data of Case study 1 referred to the FU (1 ha of decontaminated soil).

Item	Units	Value
i. Biomass cultivation & harvesting		
Area	ha	1
Seeds	kg	930
Diesel (labor machinery)	kg	$7.30 \cdot 10^3$
Nitrogen fertilizer	kg	$5.58 \cdot 10^3$
Phosphate fertilizer (P_2O_5)	kg	$1.24 \cdot 10^4$
Potassium fertilizer (K_2O)	kg	$6.20 \cdot 10^3$
ii. Biomass conditioning & transport		
Produced biomass (DM)*	kg	$7.48 \cdot 10^5$
Pb extracted	kg	$3.58 \cdot 10^2$
Electricity	kWh	$8.30 \cdot 10^5$
Diesel (transport)	kg	$2.80 \cdot 10^3$
iii. Biomass use		
A) Disposal		
Contaminated biomass (landfill disposal)	kg	$7.48 \cdot 10^5$
B) Energy recovery		
Biomass formula		$C_7H_{11}O_6$
Compressed air (biomass drying and cogeneration)	Nm^3	$9.86 \cdot 10^6$
Heat	kWh	$1.85 \cdot 10^6$
Electricity	kWh	$1.29 \cdot 10^6$
CO_2 emissions (off-gas)	kg	$1.20 \cdot 10^6$
Ash (landfill disposal)	kg	$3.58 \cdot 10^2$
Diesel (transport for ash disposal)	kg	1.33

* DM: dry matter.

2.3.2. Case study 2: soil washing

This process consists of removing the Pb contained in the soil by using chemicals, firstly HCl to solubilize Pb and then, NaOH for the subsequent precipitation. This process was simulated in SuperPro Designer 9.5. For this purpose, the process described by Kim et al. (2013) was taken as a reference assuming a treatment capacity of 10 m^3 h^{-1}. The process flowsheet of this treatment is shown in the supplementary material (Fig. S2), and the simulation results (chemicals and energy consumption) are summarized in Table 2 referred to the FU. The steps involved are the following:

i. *Excavation*: The soil was excavated by using an excavator and a skid steer at 20 cm depth in order to assure the collection of Pb.

ii. *Transport:* The contaminated soil is transported to the treatment plant. As these facilities do not exist, a distance of 15 km was supposed (half the distance between the contaminated area and the landfill).

iii. *Pretreatment*: The contaminated soil is separated regarding its particle size. With this purpose, the soil is passed through a vibratory wet separator in which the particles are separated into two streams: one containing particles within the range 1.5–4 mm and the other one formed of particles <1.5 mm. The fraction containing particles within the range 1.5–4 mm is dewatered by centrifugation and treated in the washing step described below. The other fraction (particle size <1.5 mm) is separated in a hydrocyclone in two streams: one containing particles with sizes <0.075 mm and the other containing particles within the range 0.075–1.5 mm. The former stream is passed to a thickener + clarification process obtaining a stream containing contaminated soil particles which are disposed in a hazardous waste landfill. The liquid fraction is treated in the wastewater treatment stage. On the other hand, the stream containing fine particles (0.075–1.5 mm) obtained before is centrifuged and then mixed with the fraction (particle size 1.5–4 mm) and conducted to the washing step. The water obtained from the centrifugation step is reused downstream.

iv. *Washing*: The stream resulting from the above step is mixed with HCl 0.2 N and thereafter centrifuged (Fig. S2, supplementary material), obtaining a clean solid (formed by particles up to 0.75 mm.) that can be further used for soil refill. The liquid fraction from this step is passed to the wastewater treatment process.

v. *Wastewater treatment*: The resulting wastewater streams from the pretreatment (hydrocyclone) and the washing step must be treated (see Fig. S2). For this purpose, the former is passed through a sequential thickening + clarification process. The solid stream free of Pb can be used is disposed as a hazardous waste is an underground security deposit. On the other hand, the stream coming from the washing step is neutralized using NaOH (0.2 N) in a neutralization tank, and the resulting effluent is mixed with the liquid streams obtained above in the thickener and clarification tanks. The resulting stream is treated in a clarification tank, yielding two streams: a clarified liquid, recycled into the process and a residual effluent which is centrifuged. The solid fraction is disposed as hazardous material in an underground deposit, and the liquid fraction is recycled to the process. The possibility of recycling the liquid streams allows reducing the consumption of freshwater.

vi. *Soil disposal*: The fraction of the soil containing Pb was disposed as hazardous material in a landfill assuming 15 km from the treatment plant. As in Case 1A, the landfill was modelled as an underground deposit adapted from Gabi Professional 2021 database.

vii. *Soil refilling*: This step consists of refilling the excavated site using fresh sand along with part of the treated soil obtained after the washing step. In this case, it was supposed that the sand was extracted from an existing quarry 30 km away from the excavated site (close to the landfill). A mini skid steer and a truck were used to charge and transport the sand, respectively. All these processes (extraction, charge and transport of sand) were taken and adapted from Gabi Professional 2021 database.

Table 2
Inventory data of Case study 2 referred to the FU (1 ha of decontaminated soil).

Item	Units	Value
i. Excavation		
Excavator	m^3	$2.00 \cdot 10^3$
Mini skid steer	m^3	$2.00 \cdot 10^3$
ii. Transport		
Diesel	kg	$4.90 \cdot 10^3$
iii. Pretreatment		
Hopper	kWh	$1.50 \cdot 10^3$
Vibrating screen	kWh	$1.78 \cdot 10^3$
Centrifugation	kWh	$3.56 \cdot 10^2$
Hydrocyclone	kWh	$2.45 \cdot 10^3$
iv. Washing		
Hydrochloric acid (20 wt% solution)	kg	$6.23 \cdot 10^3$
Washing tank (0.2 N HCl solution)	kWh	$3.46 \cdot 10^3$
Centrifugation	kWh	$3.88 \cdot 10^2$
v. Wastewater treatment		
Sodium hydroxide	kg	$2.96 \cdot 10^4$
Neutralization tank (0.2 N NaOH)	kWh	$2.36 \cdot 10^3$
Thickening	kWh	$1.36 \cdot 10^2$
Clarification	kWh	$2.32 \cdot 10^2$
Centrifugation	kWh	$1.36 \cdot 10^2$
vi. Soil disposal		
Disposal, hazardous waste	kg	$1.14 \cdot 10^6$
vii. Soil refilling		
Raw sand	kg	$1.80 \cdot 10^6$
Diesel (transport)	kg	$6.73 \cdot 10^3$
Mini skid steer	m^3	$2.00 \cdot 10^3$

2.3.3. Case study 3: excavation and landfill deposition

Inventory data of the selected treatments for this case are summarized in Table 3. In this process, the following steps were considered, as shown in Fig. 1:

i. *Excavation*: This process consists of extracting the contaminated soil by excavation. For this purpose, an excavator and a mini skid steer were used. Both processes were taken from Gabi Professional 2021 database and adapted to the functional unit, obtaining the values summarized in Table 3.

ii. *Soil deposition*: The contaminated soil is charged and transported to the same landfill as in Case 2. The transport was carried out by truck, and the landfill was modelled, considering the contaminated solid as a hazardous waste due to the presence of Pb. These processes were also adapted from Gabi Professional 2021 database.

iii. *Soil refilling*: The final step of the process consists of refilling the excavated site by using fresh sand from a quarry located 30 km away. The same amount of fresh sand as excavated soil (3000 tons) was used to carry out this task. A mini skid steer and a truck were used to charge and transport the sand, respectively, from the quarry to the excavated site.

2.4. Environmental Impact Assessment

The evaluation of environmental impacts was performed by using the mid-point approach. The impact categories were selected from those recommended by the ILCD handbook (European Commission, 2010), taking into account the most typically used for LCA applied to soil bioremediation (Visentin et al., 2019). Greenhouse gas emissions (quantified through 100 year Global Warming Potential category, GWP), acidification and eutrophication potentials (AP and EP, respectively), as well as toxicity-related impacts: ecotoxicity and human toxicity potentials (ETP and HTP, respectively). These selected categories were quantified by using CML 2001-Aug.2016 methodology. Apart from that, Cumulative Energy Demand (CED) was calculated to quantify the direct and indirect primary energy use throughout the life cycle (Huijbregts et al., 2010). Both fossil and renewable energy were considered.

Table 3
Inventory data for Case study 3 referred to the FU (1 ha of decontaminated soil).

Item	Units	Value
i. Excavation		
Excavator	m^3	$2.00 \cdot 10^3$
Mini skid steer	m^3	$2.00 \cdot 10^3$
ii. Soil disposal		
Diesel (transport)	kg	$11.20 \cdot 10^4$
Disposal, hazardous waste to underground deposit	kg	$3.00 \cdot 10^6$
iii. Soil filling		
Raw sand	kg	$3.00 \cdot 10^6$
Diesel (transport)	kg	$11.20 \cdot 10^4$
Excavator	m^3	$2.00 \cdot 10^3$
Mini skid steer	m^3	$2.00 \cdot 10^3$

3. Results and discussion

Table 4 includes the results obtained for the different impact categories considered in this work, including the avoided impacts by electricity and heat generation in Case 1B. As can be observed, Case 1B shows the lowest impact values in all categories. This finding is remarkable since cogeneration of biomass requires additional energy and material inputs (air and heat for biomass drying and combustion) but these requirements are balanced by the avoided impacts yielded by energy production. These results are in agreement with other works on the removal of heavy metals from soils using other species plants (Vocciante et al., 2021).

3.1. Impact contribution analysis

In this section, the contribution of each step to the overall impact in the studied cases was carried out.

3.1.1. Case study 1: phytoextraction treatment

In this section, quantification of environmental impacts of Case 1 was carried out considering two options for biomass disposal: security deposit (Case 1A) and energy recovery (Case 1B). Fig. 2 depicts the relative contribution of the different steps with respect to the overall results shown in Table 4. As can be observed, disposal of contaminated biomass in Case 1A is the most important contributor to all impact categories (ranging from 50 to 70%) due to the high amount of contaminated biomass to be treated, in agreement with literature (Vocciante et al., 2021). On the other hand, biomass conditioning and transport step contribute to all the impact categories by 30–40% in this scenario because of the energy required to biomass conditioning (screen vibration) before cogeneration step. Regarding Case 1B, the biomass conditioning and transport step show the highest contribution to all the impact categories except CED and GWP, mainly affected by the cogeneration step. Concerning CED, the main contributor is the compressed air used for drying and combustion of the biomass in the boiler. On the other hand, the high contribution of cogeneration to GWP is due to CO_2 emissions resulting from biomass combustion. These categories are mainly affected by biomass cogeneration because of material and energy consumption (compressed air and heat needed for drying step before cogeneration) as well as CO_2 emitted during this process, respectively. Concerning cultivation step, it can be observed in Fig. 2B, how eutrophication category (by 30%) and toxicity related impacts (HTP and ETP, by 20%) are the most affected categories due to harmful emissions (mainly heavy metals) generated by fertilizer production. It must be noticed the negligible contribution of the disposal step in this scenario, which is directly related to the dramatic reduction of hazardous waste produced in the cogeneration step (only ashes containing Pb).

As above mentioned, the use of cogeneration is superior to landfill disposal because of the avoided impacts yielded by the energy production. Fig. 3 depicts the comparison of overall impacts in Cases 1A and 1B, including the avoided ones, calculated according to the Spanish electric production mix and natural gas combustion for electricity and heat production, respectively. Net impacts were calculated as the difference between the overall and avoided impacts.

As can be seen (Fig. 3), Case 1B shows lower gross impact values in all categories, except for GWP and CED, in which cogeneration is the main contribution. As above commented, this result is directly related to the impacts generated by the use of compressed air and heat for biomass drying (prior to cogeneration), which make energy needs increase, enlarging CED of the process. On the other hand, CO_2 emitted from biomass cogeneration is the main the responsible of GWP rise. For the rest of categories, biomass conditioning and transport is the most important contributor, mainly due to the energy consumed in both steps.

Table 4
Overall impacts for the case studies referred to the FU (1 ha of decontaminated soil).

Impact category	Case 1A	Case 1B	Case 2	Case 3
CED (MJ)	$1.80 \cdot 10^7$	$3.98 \cdot 10^6$	$7.88 \cdot 10^6$	$1.31 \cdot 10^7$
GWP (kg CO_2-eq)	$6.15 \cdot 10^5$	$1.63 \cdot 10^5$	$3.55 \cdot 10^5$	$4.75 \cdot 10^5$
AP (kg SO_2-eq)	$2.75 \cdot 10^3$	$4.77 \cdot 10^2$	$1.37 \cdot 10^3$	$2.24 \cdot 10^3$
EP (kg PO_4-eq)	$1.28 \cdot 10^3$	$1.44 \cdot 10^2$	$7.40 \cdot 10^2$	$1.27 \cdot 10^3$
HTP (kg 1,4 DCB-eq)*	$2.27 \cdot 10^5$	$-1.02 \cdot 10^4$	$1.39 \cdot 10^5$	$1.94 \cdot 10^5$
ETP (kg 1,4 DCB-eq)*	$2.86 \cdot 10^3$	$2.48 \cdot 10^2$	$2.26 \cdot 10^3$	$1.98 \cdot 10^3$

* DCB: dicholorobenzene.

By comparing the net impact values of Cases 1A and 1B, it can be observed that in all categories the former is superior to the former. In this sense, CED net impact of Case 1B is 84% lower than for Case 1A, enhancing remarkably overall energy requirements of the process. By considering the contribution of electricity and heat to avoided impacts, it can be observed that the former is the main contributor, despite larger amount of heat is produced. The reason for this result is that the energy avoided per MJ of electricity produced is higher (2.55 MJ) than in the case of heat (natural gas, 1.41 MJ), supporting the obtained results.

With regard to GWP impact, it is clearly improved by avoided impacts, achieving a reduction with respect to Case 1A by 80%. For this category, the avoided impact contribution is clearly dominated by the heat production due to both the larger amount of heat produced as well as the higher avoided CO_2 emissions per MJ of energy produced (0.078 kg CO_2 eq MJ^{-1}) compared to those from the Spanish electricity mix (0.053 kg CO_2 eq MJ^{-1}), according to the impacts of these processes obtained by LCA software. The benefits of using biomass for energy purposes instead of landfill disposal after phytoextraction, in terms of CO_2 emissions, were also reported by Vocciante et al. (2019). In this sense, carbon footprint values obtained in our work and those reported by Vocciante et al. (2019) were of the same order, although somewhat higher in our case because of the larger amount of biomass needed. This comparison must be taken with caution and only at a qualitative level as the biomass used by these authors (*Lupinus albus*, *Brassica juncea* and *Helianthus annuus*) as well as the characteristics of the polluted soil were different.

Avoided impacts positively affect the net impact value for both AP and EP categories in Case 1B. Thus, the net impact values obtained are nearly 83% and 89% lower, respectively, than in Case 1A, showing the superiority of biomass cogeneration compared to landfill disposal. By analyzing the avoided impacts within AP and EP categories, it can be observed that they were larger for electricity than for heat, despite the production of the former by cogeneration is lower. In the case of AP impacts, this result can be explained because of the higher inorganic emissions (SOx) generated by the Spanish electricity mix ($2.96 \cdot 10^{-4}$ kg SO_2 eq MJ^{-1}) in comparison to natural gas ($7.1 \cdot 10^{-5}$ kg SO_2 eq MJ^{-1}), mainly due to coal contribution. Regarding EP avoided impacts, a similar trend was obtained since the emissions of phosphate-eq per unit of energy produced are remarkably larger for electricity ($6.78 \cdot 10^{-5}$ kg phosphate eq MJ^{-1}) than for natural gas ($1.81 \cdot 10^{-5}$ kg phosphate eq MJ^{-1}).

Finally, biomass cogeneration reduces the net impact value for toxicity related impacts (HTP and ETP). In this sense, avoided impacts due to the biomass cogeneration achieve reductions of 100% and 91% for HTP and ETP, indicating the superiority of Case 1B compared to Case 1A. By analyzing the contribution of electricity and heat production to avoided impacts, it can be observed (Fig. 3) that the former is clearly higher, although less electricity than heat is produced by cogeneration. This result is explained due to the higher amount of harmful emissions avoided per MJ of electricity produced. In this sense, avoided emissions by the Spanish electricity mix for HTP ($2.31 \cdot 10^{-2}$ kg DCB eq MJ^{-1}) and ETP ($3.32 \cdot 10^{-4}$ kg DCB eq MJ^{-1}) are larger compared to those from heat ($5.30 \cdot 10^{-3}$ kg DCB eq MJ^{-1} for HTP and $7.23 \cdot 10^{-5}$ kg DCB eq MJ^{-1} for ETP). The responsible for this difference is the contribution of fossil resources in the Spanish electricity mix used in this work.

According to the results above explained, biomass energy valorization by cogeneration is clearly superior to security landfill disposal because avoided impacts from electricity and heat production affect positively the overall footprint of the treatment. This result is in agreement with other works reported in the literature for phytoremediation of heavy metals using different species of plants (Todde et al., 2022; Vocciante et al., 2021).

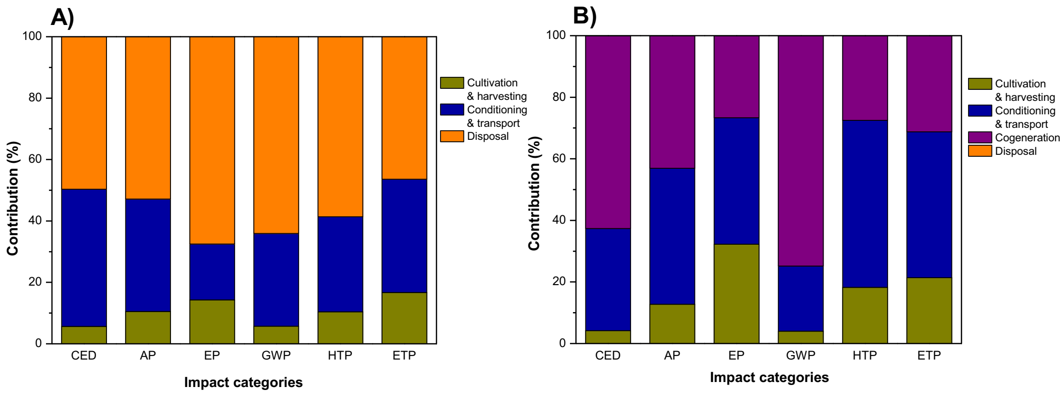

Fig. 2. Relative contribution of the different steps to the overall impacts: (A) Case study 1A and (B) Case study 1B.

3.1.2. Case study 2: soil washing

Fig. 4 shows the relative contribution of the steps involved in the process to the selected impact categories. It can be observed how soil disposal is the most important contributor to all categories (55–70%), except to ETP. The main reason is the large amount of soil to be disposed as hazardous material (~50% of the treated soil) in the security landfill. Washing is the second contributor in order of importance in all categories (15–40%), being the first one for ETP (~60%). The main responsible for this high impact is the production of hydrochloric acid used to solubilize the Pb contained in the soil, which production implies a large amount of energy as well as emissions affecting GWP (~15%), AP and EP categories (~20–25%, respectively). Regarding HTP and ETP, soil washing is the

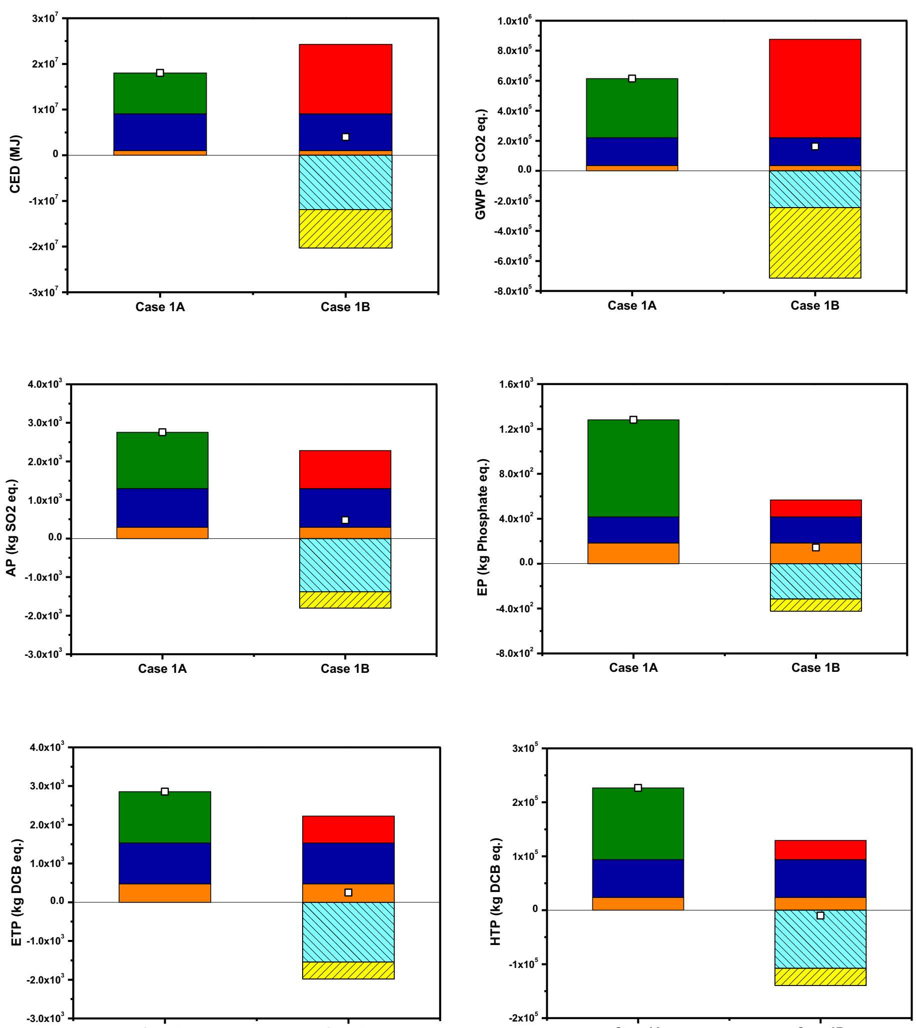

Fig. 3. Comparison of the calculated impacts between Case 1A and 1B. Solid bars: gross impact (orange: cultivation & harvesting; blue: conditioning & transport; red: cogeneration; green: disposal); striped bars: avoided impacts (cyan: electricity; yellow: thermal energy); white square: net impact.

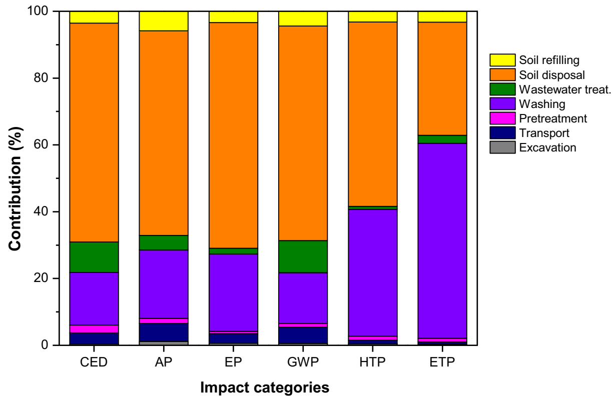

Fig. 4. Relative contribution of the different steps to the overall impacts for Case study 2.

main contributor (~40% and 60%, respectively) because of the emissions of heavy metals by hydrochloric production, which for the latter category even exceeds the soil disposal contribution. Finally, wastewater treatment and soil refilling contribute by 5–10% to the selected categories due to the impacts related to NaOH production and the impacts produced by the extraction activities of sand, respectively.

LCA results for soil washing treatment indicate that soil disposal and production of HCl are the main responsible of the environmental impacts studied in this work.

3.1.3. Case study 3: excavation and landfill deposition

The relative contributions to the studied impact categories are depicted in Fig. 5. In this case, soil disposal is the most significant contributor (>90%) to all the impact categories. This result could be expected since, in this case, the whole contaminated soil is directly treated in a security landfill (3000 tons). By comparing the relative contribution of the soil disposal step to the overall impact concerning Case study 2, it is observed that this contribution is lower than in Case study 3, since the amount of soil to be disposed is minor (1140 tons vs 3000 tons). Regarding the refilling step, the relative contribution in Case 2 and Case 3 is slightly higher for the latter. These results could not be expected since the amount of sand is clearly higher in Case 3 (3000 tons vs 1800 tons). However, this step shows much lower contributions in both cases than soil disposal, as this includes materials and operations that generate huge impacts. This fact leads to a similar relative contribution value in both cases.

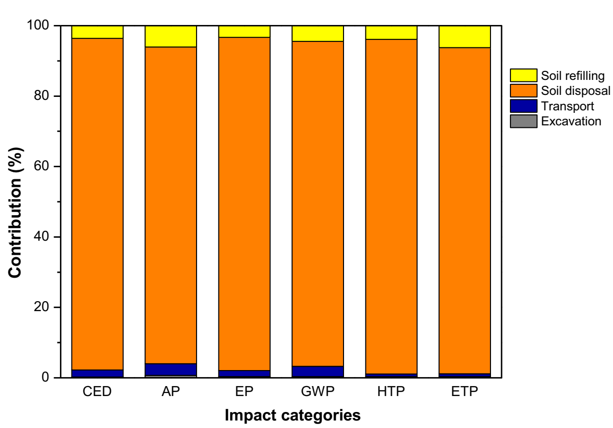

Fig. 5. Relative contribution of the different steps to the overall impacts for Case study 3.

3.2. Comparison of the different treatments

In order to check the environmental performance of the studied treatments, they were compared and the results depicted in Fig. 6. The most environmentally friendly treatment in all categories is phytoextraction with energy recovery (Case 1B). On the contrary, Case 1A and Case 3 present the highest impacts in all categories because of the large amount of biomass or soil that has to be disposed as hazardous waste in a security landfill.

By analyzing the effect of the biomass disposal option, it can be observed how biomass cogeneration clearly enhances the environmental footprint of Pb-phytoextraction by *F. arundinacea*. In this sense, reductions ranging between 70 and 100% were obtained for all impacts. As above explained, the reduction of the soil disposed in the landfill, as well as the avoided impacts due to the energy recovery from biomass are the factors that make superior the use of biomass with energy ends. This result agrees with the results obtained by Vocciante et al. (2021), which reported the superiority of the energy use from biomass over disposal in terms of GWP impact (the rest of impacts were not analyzed by these authors).

By comparing Case 1B with the rest of the treatments, it can be observed that it is superior in all categories, mainly due to the avoided impacts related to the produced energy (electricity and heat). Thus, reductions ~20% and ~50% were obtained for CED with respect to Case 2 and Case 3, respectively. In terms of GWP, Case 1B yields reductions above 30% and 70% compared to Case 2 and Case 3. A similar trend was reported by Vocciante et al. (2019 and 2021) for phytoextraction + energy recovery using *Lupinus albus*, *Brassica juncea*, and *Helianthus annuus* to remediate a Pb-contaminated soil. Concerning AP and EP, Case 1B also shows better environmental performance than Cases 2 and 3, yielding impact reductions by 33% and 64%, respectively. By analyzing HTP, the reductions achieved by Case 1B are 61% and 85% with regard to Cases 2 and 3, respectively. Finally, Case 1B shows reductions by 70% and 60% compared to Cases 2 and 3, respectively.

Overall, phytoremediation of Pb using *F. arundinacea* is superior to soil washing and excavation treatments. Furthermore, energy recovery from biomass is superior to landfill disposal, since the former option allows reducing the biomass to be disposed as well as avoiding impacts related to energy production. Consequently, the environmental footprint of the process is enhanced.

Hence, phytoextraction and further energy recovery from the biomass is an advantageous option as compared to traditional physical and chemical treatments for highly Pb-contaminated soils, despite its longer duration.

4. Conclusions

LCA methodology was applied to study the environmental performance of phytoextraction using *Festuca arundinacea* as well as other traditional treatments to remediate a Pb-contaminated soil. The most important impacts using phytoextraction + landfill were caused by the large amount

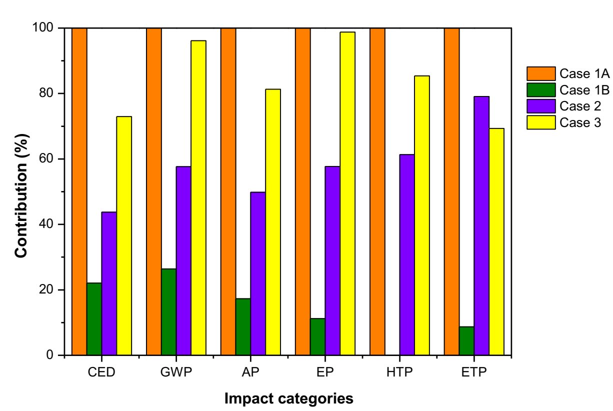

Fig. 6. Normalized comparison for the case studies referred to the functional unit.

of biomass disposal. On the other hand, biomass conditioning and cogeneration steps were the most important contributors to the environmental impacts because of energy consumption and CO_2 emissions, respectively. The combination of phytoextraction + energy recovery reduces the environmental impacts in all the studied categories compared to direct biomass disposal due to the avoided impacts by electricity and heat production. Regarding traditional methods, the production of hydrochloric acid is the most important contributor to the environmental impacts of soil washing treatment. In contrast, soil disposal presents the highest contribution in the case of excavation + landfill scenario. Finally, phytoextraction (including biomass disposal or energy recovery) was compared with physical (soil excavation) and chemical (soil washing) treatments, obtaining remarkable reductions when using biomass to produce energy in all the studied impacts. Accordingly, the use of phytoextraction using *F. arundinacea* along with energy recovery, despite its longer duration, is an attractive option for remediation of highly Pb-contaminated soil from an environmental point of view.

CRediT authorship contribution statement

Juan J. Espada: Conceptualization, Writing – original draft. **Rosalía Rodríguez:** Conceptualization, Writing – original draft. **Vanessa Gari:** Conceptualization, Investigation. **Pablo Salcedo-Abraira:** Formal analysis, Writing – original draft. **Luis Fernando Bautista:** Supervision, Writing – original draft.

Declaration of competing interest

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.156675.

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