Can the green energies improve the sustainability of electrochemically-assisted soil remediation processes?

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Abstract

The green powering of electrochemically-assisted soil remediation processes had been strongly discouraged. Low remediation efficiencies have been reported as a consequence of the reversibility of the transport processes when no power is applied to the electrodes, due to the intermittent powering of renewable sources. However, it has been missed a deeper evaluation from the environmental point of view. This work goes further and seeks to quantify, using life cycle assessment tools, the environmental impacts related to the electro-kinetic treatments powered by different sources: grid (Spanish energy mix), photovoltaic and wind sources. The global warming potential and the ozone depletion showed higher environmental impacts in case of using green energies, associated with the manufacturing of the energy production devices. In contrast to that, results pointed out the lowest water consumption for the treatment powered with solar panels. The huge water requirements to produce energy, considering a Spanish energy mix, drop the sustainability of this powering strategy in terms of water footprint. Regarding toxicities, the pollutant toxicity was highly got rid of after 15 days of treatment, regardless the powering source used. Nevertheless, the manufacturing of energy and green energy production devices has a huge impact into the toxicity of the remediation treatments, increasing massively the total toxicity of the process, being this effect less prominent by the electro-kinetic treatment solar powered. In view of the overall environmental impact assessed, according to mid and endpoint impact categories, it can be claimed that, despite the high energy requirements and affectation to the global warming potential, the use of solar power is a more sustainable alternative to remediate polluted soils by electrochemical techniques.

Keywords

Life cycle assessment; soil remediation; electrochemical; environmental impacts; photovoltaic power; wind power.

Highlights

- EASRP sustainability is influenced by the nature of its powering source.

- The use of a wind powering leads to important environmental impacts

- Soil toxicity noticeable drops after an EASRP, regardless of the powering source.

- EASRP sustainability strongly influenced by manufacturing of green powering devices.

1. Introduction

Removal of hazardous contaminants from polluted soils is one of the topic of major relevance nowadays (Lacasa et al., 2019). Diffusion mechanisms may transport the pollution contained in soil and jeopardize the quality of water reservoirs which does not only affect the ecosystems but also the human health (Rodrigo et al., 2014; Fatin et al., 2019). Because of that, a large variety of soil remediation technologies are being developed in the last decades (Gomez et al., 2010; Pardo et al., 2016; Zhao et al., 2016), being some of them at very high technology readiness levels (TRLs), while other are still at early stage of research. Among them, it is worth mentioning that electrochemically assisted soil remediation processes (EASRP), in which an electric field is applied to electrodes placed in soil producing many chemical, electrochemical, electrokinetic and thermal changes, reduce the impact of the pollutants contained in the soil, depleting them from soil or transforming them into less hazardous species (Bocos et al., 2014; Buchireddy et al., 2009; Fonseca et al., 2012; Hamdan et al., 2014; Millan et al., 2020; Reddy et al., 2011; Sun et al., 2017; Virkutyte et al., 2002). Furthermore, it is important to note that this technology has reported promising remediation efficiencies at large scales. Risco et al. (Risco et al., 2016a; Risco et al., 2016b; Risco et al., 2016c) and Lopez-Vizcaino et al. (Lopez-Vizcaino et al., 2016a, b) stated novel statements in this field using pilot (175 dm^3) and prototype (32 m^3) plants, respectively. Those works exposed that different mechanisms may dominate the pollutant dragging regarding the scale of the remediation setup. While electrokinetic mechanisms control the remediation processes carried out at lab-scale, the thermal processes predominate in the treatments at large-scale.

Up to few years ago, the main target of those developing technologies was to reach high efficiencies in the remediation of soils, that is, to remove fast and completely the pollutants contained, preventing their spreading (Cameselle and Reddy, 2013; Lu and

Yuang, 2009; Reddy et al., 2011). However, in the recent years sustainability concepts have arisen and now, it is not only required to develop efficient technologies in the removal of pollutants, but it is also relevant that these technologies are capable of minimizing environmental impacts (López-Vizcaíno et al., 2019; Millan et al., 2020). It would not make sense to perform remediation treatments that bring out higher environmental risks that the impact to be recovered. According to that, the novel term "green remediation" emerged (EPA, 2008). It states the bases to perform cleanup actions, minimizing their environmental impacts.

These actions are essential to prevent several of the warnings that currently we are facing due to the climate change consequences (in the context of the circular economy). At this point, life cycle assessments (LCAs) have arisen as an important tool to evaluate the sustainability of processes, products and services. Currently, this methodology is being applied to evaluate the sustainability of many environmental technologies including soil remediation techniques (da S Trentin et al., 2019; Lemming et al., 2012; Lemming et al., 2009; Vocciante et al., 2016, ; Vocciante et al., 2019).

Regarding EASRP, many relevant information for the design of full-scale processes have been obtained in the recent years. Results show that a complex set of processes influence the recovery of a polluted soil, the dragging of species does not only depend on the process operation conditions (electric field, electrodes placement…) but also, and, very importantly, on soil and pollution characteristics (López-Vizcaíno et al., 2017a, b). Furthermore, it is important to note that the higher the experimental setup size, the higher the operational costs, being the powering costs the most relevant (López-Vizcaíno et al., 2019). Considering that the power consumption of those treatments is one of the most important economic costs, the nature of that energy may also influence the environmental cost of the process. Thus, powering EASRP with renewable energy has been pointed out as a promising alternative in the search of greener processes (Ganiyu et al., 2020). In addition, keeping in mind that the electrochemical technologies are powered by direct current (DC), their coupling with green energies could be the easiest and the most environmentally friendly way to operate those remediation processes. Even though a green powering could seem a sustainable alternative to the traditional grid powering, results obtained have not been always positive (Souza et al., 2016a; Souza et al., 2016b). Thus, many works have concluded that the direct application of solar or wind energies without using energy storage devices leads to very inefficient processes from the viewpoint of pollutant removal, because of the reversion of the transport processes when no energy is supplied to an electro-remediation system. The low efficiencies reported by green powered electrochemical technologies could be explained because of the transport of pollutants or carriers (such as surfactants) is not only interrupted but reversed overnight (when powering with PV panels) o in not-windy periods (when powering with wind turbines).

Anyhow, despite these lower efficiencies, there is still a doubt regarding the most sustainable powering considering non only energy but also all the inputs and outputs of the process. In a previous work of our group, it has been demonstrated that for electrolytic treatment technologies of liquid waste, the sustainability of processes noticeably increases when a photovoltaic (PV) powering is applied (Fernández-Marchante et al., 2021). In that case, processes are mainly irreversible, and the lack of powering do not produce any meaningful reversion in remediation mechanisms.

Considering the relevant results reached under the green treatments of wastewater effluents in terms of sustainability, this work is aimed at evaluating the environmental risks of powering EASRPs using the power grid (Spanish energy mix) or a green powering (solar photovoltaic panels or wind turbines). It is worth mentioning that despite green powered EASRPs reported lower efficiencies by the same time of treatment, the LCA studies could shed light of an opposite trend in terms of sustainability. For that reason, it is key to assess the suitability of a treatment not only in terms of remediation efficiency but also in terms of sustainability. Thus, to strike a balance between efficiency and sustainability must be the most important fact to be considered before performing a remediation treatment.

According to that, until now, many research groups have focused their studies on the sustainability of different energy sources (Goel et al., 2009; Itten et al., 2012; Turconi et al., 2013; Wang et al., 2019). Many of those studies confirm that green energies are essential to reduce the impact related to the production of energy by means of fossil fuel. Furthermore, they confirm that wind power has lowest environmental impacts than PV power, considering the same production of energy. Despite those studies claim against the use of energy coming from fossil fuels due to their huge environmental risks per unit of energy, in this case, the environmental risks of an electrochemical remediation will not only directly depend on the nature of the energy supplied by the treatment but also on the total energy consumption and the level of remediation reached by a specific period of time. Consequently, it is required to evaluate the overall impacts of the EASRP powered by different sources before performing statements according to its sustainability. This LCA may work as a tool to make decision according to the implementation of remediation techniques, which would allow to reduce the environmental and economic impacts of a recovery treatment.

To do that, experimental data previously obtained by our research group were used to take into account all the input and output of the systems. SimaPro 9.0 was used as software tool and Ecoinvent 3.3 as data base to carried out the inventory of the equipment of each remediation setup. To determine the most meaningful impacts discerned from these remediation analyses, AWARE, USEtox, IPPC and ReCiPe methodologies were used to quantify the environmental burden into 5 midpoint (water footprint, global warming potential, ozone layer depletion, human toxicity, freshwater ecotoxicity) and 17 endpoint impact categories.

2. LCA methodology

2.1. Methods

To quantify the environmental impact related to the EASRP of a polluted soil, SimaPro 9.0 was used as software tool. To determine the most meaningful impacts discerned from these remediation analyses, the ReCiPe method was implemented. To improve the quality of the results, IPCC, AWARE and USEtox methods were used to complement the data obtained in terms of global warming, water footprint and toxicology, respectively (Fernandez-Marchante et al., 2020; Fernández-Marchante et al., 2021a; Fernández-Marchante et al., 2021b; Zhang et al., 2019). Those methodologies allow to allocate a value to the different mid-point and end-point impact categories studied: fossil depletion, metal depletion, natural land transformation, urban land occupation, agricultural land occupation, marine ecotoxicity, terrestrial ecotoxicity, freshwater eutrophication, terrestrial acidification, climate change ecosystems, ionising radiation, particulate matter formation, photochemical oxidant formation, human toxicity, ozone depletion and climate change human health.

2.2. Goal and scope

The LCA will address the environmental impacts of EASRPs powered by different energy sources. To perform this study and to determine the inputs and outputs of the treatment, experimental results previously reported by our group were used (Souza et al., 2016a; Souza et al., 2016b). Those studies carried out the EASRP of a soil polluted with 150 mg 2,4-D per kg of dry soil using a bench scale plant and under different powering strategies:

grid and direct random connection to photovoltaic panels and a wind turbine, without using energy storage devices. This study seeks to evaluate the environmental impacts associated with a soil remediation treatment from cradle to grave. It is worth mentioning that the reuse or recycling of the components of the studied setups have not been considered. Thus, the analyses bring out the environmental risks from the cradle to the end of the lifespan of the remediation system.

Figure 1 shows a schematic flowsheet of the treatment that helps to define the boundaries of the systems. The foremost inputs of the remediation process are the energy powered, the polluted soil and the flushing fluid used for the dragging of the pollutant from the soil to the wells. The outputs are the treated soil, the flushing effluent that should contain the pollutant originally presented into the soil and the gas flow emitted during the EASRPs, which can drag part of the pesticide and lead to polluting air.

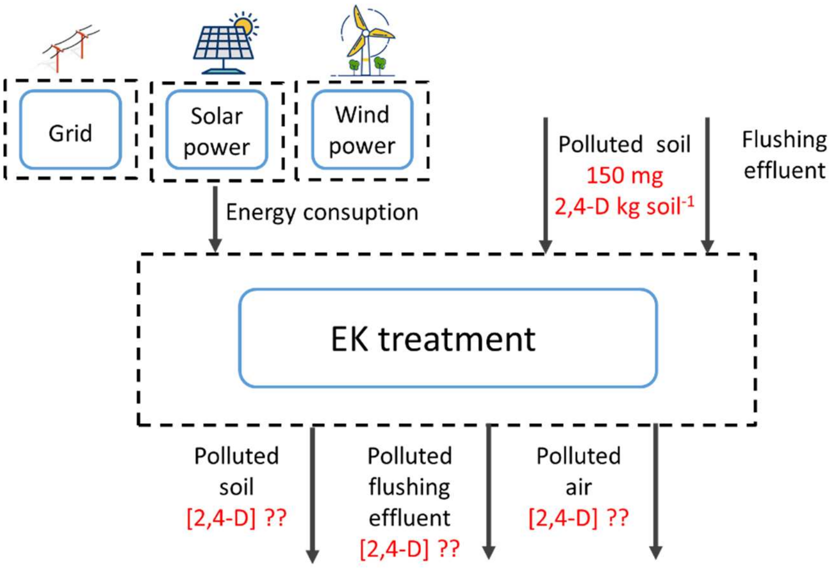

Figure 1. Schematic flowsheet of the treatment.

The EASRP powered directly by the grid, worked at potentiostatic mode, providing a constant potential gradient of 1.0 V cm^{-1}. Conversely, the direct random connections between the EASRP and the green energy production devices (PV plant and the wind turbine) showed a fluctuating powering according to the weather conditions. Furthermore, it worth mentioning the green energy production devices were not designed according to the EASRP which could lead to an inefficient powering. Thus, considering this fact, the efficiency and sustainability of those treatments could be directly related to the fluctuating power production of green energies.

The treatments were run for 15 days in all the cases, regardless the level of remediation reached. Figure 2 summarizes the electro-osmotic and evaporation flows and the remained pollutant in soil after 15 days of electrokinetic treatment for each powering strategy studied. The electroosmotic flows are determined by Darcy`s law and directly depended on the electric potential applied to the treatment (Alshawabkeh et al., 1993). Contrary to expectations, the EASRP powered by the grid showed the highest electroosmotic flows and consequently the highest dragging of pollutant to the wells despite the lowest charge supplied to the soil matrix during 15 days of treatment, 4.3 Ah kg^{-1}. In this case, a 90.2 % of pesticide was transported from the soil matrix to the wells after 15 days of EASRP. Conversely, the EASRP powered by wind energy reported the worst remediation despite the highest charge passed throughout the soil matrix in this case, 49.2 Ah kg^{-1}. Under those operational conditions, a 49.2 % of the initial pesticide was removed from the soil.

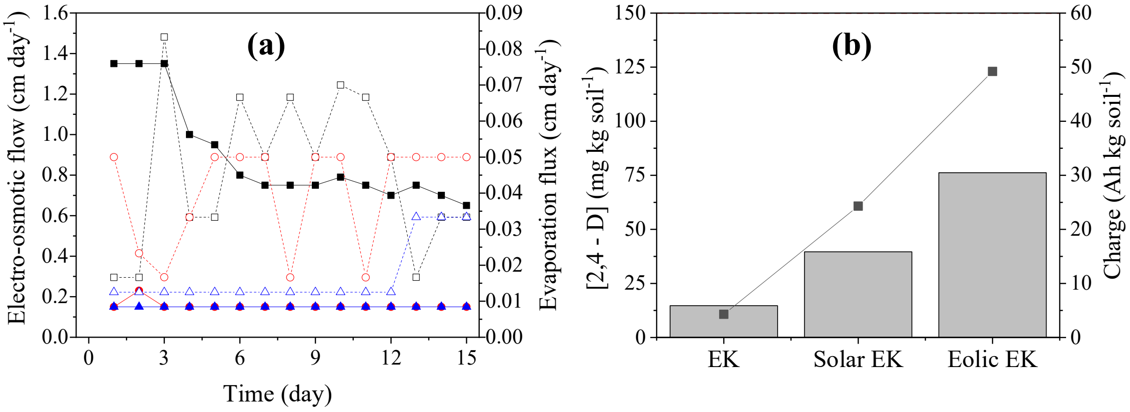

Figure 2. a) Electro-osmotic (full symbol) and evaporation (empty symbols) flows during the electrokinetic treatments powered by grid (■), solar panels (●) and a wind turbine

(▲). b) 2,4-D concentration into the soil matrix and charge passed after 15 days of remediation treatment. Initial 2,4-D concentration: 150 mg kg soil^{-1}.

The fluctuating current supplied by renewable energies points out a lower overall performance of these remediation techniques. Those results can be explained by means of the transport of species produced when the electric field is applied between electrodes. The dragging of pollutant to the wells decreases or reverses when the powering drops or ceases (Millan et al., 2020), respectively. This effect can be particularly important in wind turbines in which the periods with no activity may be longer and more distributed than in a PV powering (when only at night there is no powering). Furthermore, peaks of power can lead to another reaction mechanisms that may jeopardize the electroosmotic, electromigration and electrophoresis flows that favor the transport of the pollutants to the wells. In addition, the low remediation noticed by the EASRPs powered by green energies could be due to a waste of energy which limits the efficiency of the transport mechanisms responsible of dragging the pollutant to the wells. Thus, it is worth mentioning that despite the direct random powering of electrooxidation treatments is not affected by the fluctuating powering provided by a PV plant or a wind turbine (Millán et al., 2018; Souza, F. et al., 2015; Souza, F.L. et al., 2015), this powering strategy significantly affects the transport of species in a soil matrix which can reduce the remediation efficiency of the EASRPs. Thus, longer treatment times must be required to reach the same remediation level when renewable energies are directly coupled to EASRPs.

On the other hand, evaporation fluxes are tied to temperature increases, because of the Joule-Thompson effect (Bradl, 2005; Reddy and Cameselle, 2009) as a consequence of high flows of current through the soil. In this case, lower evaporation flows were noticed during the experimental tests, being those values almost negligible regarding the electroosmotic flows.

Even though the removal of green powered EASRPs seems to be not as efficient as the traditional grid powered treatment, the overall performance of an EASRP can bring out new insights if the environmental impacts of these treatments is considered. The environmental risks related to the production of the energy by means of the three proposed cases can reveal interesting results with the aim of choosing the most suitable remediation technique, striking a balance between remediation efficiency and environmental impact aiming to the most sustainable recovery of natural resources.

In view of the first approaches noticed above, the following assumptions were set up to carried out the LCAs.

- The oxidation of pesticide to CO_2 was not considered. The use of graphite as electrodes and the large distance between them, reduce the possibility of arising oxidation reactions during the treatment, because of oxidation of water and self-combustion of graphite are produced in an easier way than oxidation of organic compounds (Risco et al., 2016a).
- Air pollution as a consequence of pollutant evaporation was not taken into account because of the low evaporation flows observed in all the cases.
- The treatment of the flushing effluent after the EASRP was not considered.

According to the previous statements and in order to perform a comparative analysis, the functional unit of the LCA must be set up. Despite the remediation analyses were carried out by a soil polluted with 150 mg of 2,4-D per kg of dry soil, results pointed out different remediation after 15 days of treatment. According to that, the functional unit of each EASR treatment was set up at the value of remediation reached at the end of treatment for each case of study, as it is detailed in Figure 2b. It important to highlight that according to the selected functional unit, the identification and quantification of the environmental impacts of this EASRP could be different because of the diverse remediation reached by each treatment and the variable power supplied by each power source. Thus, if a total recovery level is selected to carry out the LCA analyses, the treatment time and the power consumption are different. Conversely, if the power supplied by the treatment is the same, the treatment time and the level of treatment fluctuate. According to these premises, it essential to evaluate the results obtained in a LCA study keeping in mind the functional unit selected in each case.

2.2. Inventory

Once selected the goal and delimited the boundaries of the process, an inventory of the three different powering modes must be performed. Ecoinvent database was selected to carried out the inventory of the EASRP, which includes the input and output of the process (equipment, reagents, energy…). Table 1 shows a review of the inventory required to perform each remediation treatment according to its powering mode.

Table 1. Life cycle inventory of the EASRPs

Setup	Equipment	Composition	Weight (Kg)	Life span (years)
EASRP setup	Electrodes	Graphite	0.37	15
	Mock-up	Polymethyl methacrylate	2.50	15
	Cable	Cable	0.02	15
	Electronic conditioner Hardware	Electronics, for control units	0.05	10
		Polyethylene, high density	0.70	
		Steel, chromium steal 18/8	0.25	
		Cable	0.05	
PV plant	Photovoltaic module (2.6 m^2)			15
Wind turbine	Windmill	Electronics, for control unit	1.50	15
		Glass fibre	8.00	
		Steel, chromium steel 18/8	16.70	
		Graphite	0.50	
		Ferrite	1.50	
		Polyethylene, high density	6.00	
		Cable	0.10	
		Polypropylene, granulate	0.50	
		Steel, low-alloyed	3.00	
		Brass	7.00	
		Bronze	0.20	

3. Results and Discussion

One of the goals of the European Union is to reach a neutral climate by 2050 (United Nations, 2020a). To achieve this target, the carbon and water footprints of processes, products or services must be zero or as low as possible. Keeping in main this global and ambitus goal and the need for a quick recovery of natural sources in order to stop or get rid of the impacts related to pollution, to assess the sustainability of new remediation techniques must be as essential as to quantify their efficiencies. LCA analyses can shed light on interesting and essential conclusions that may be key to determine the best suitable remediation technique to treat a polluted soil. According to that, the environmental risks of an EASRP powered under different strategies were assessed with the aim of determining the most suitable way to run this electrochemical process.

In order to quantify the carbon footprint of those processes, one of the most important parameters to be evaluated is the greenhouse gases impacts, consequence of the production, use and end-of-life of a process, product or service. Among the wide variety of impact categories, the Global Warming Potential (GWP) outlines the environmental impact related to the greenhouse emissions, which are the main causes of the climate change. Considering the noticeable interest of minimizing these emissions, the development of novel sustainable techniques is essential. Therefore, to quantify the impact of those emissions the GWP of the different EASRPs was evaluated and plotted as equivalents kg of carbon dioxide per equivalent unit as shown in Figure 3. Data shows higher GWP impacts by the EASRP powered by wind turbines and solar panels. Results notice that 162 and 104 g of CO_2 equivalents kg^{-1} soil were emitted to the atmosphere during 15 day of EASR treatment powered with those renewable energies, respectively. Contrary to expectation, the remediation treatment powered by the grid exposed the lowest GWP value and only 49 g of CO_2 equivalents kg^{-1} soil were discharged under a traditional EASRP. These surprising results can be explained by the large amount of energy supplied to the wind powered EASRP during the 15 days of treatment, 10 times higher than by the grid powering mode. The direct connection of a green energy production device without a detailed design according to the treatment that is going to be powered, may lead to an uncontrolled powering and consequently to an inefficient and non-sustainable remediation.

Those results also show a huge environmental impact related to the manufacturing of the green energy production systems, being higher by the wind turbine.

In contrast to the results attained by other authors, wind turbines showed higher environmental impacts that PV panels. Oğuz et al. and Zhong et al. reported that the manufacturing of wind turbines has lower greenhouses emissions than the PV panel production considering a LCA “from cradle to grave” that includes the disposal stage (Oğuz and Şentürk, 2019; Zhong et al., 2011). The huge amount of material that may be recycled from a wind turbine reduces its global impact. In addition, it is important to take into account that this study consider the use of green powering sources for a detailed period of time and not for a specific power production. Thus, the differences between the three technologies can be explained by means of the energy supplied by these green energy production devices to the EASRP during the 15 days of treatment. The smallest charge supplied to the EASRP powered by the grid (4.33 Ah kg soil^{-1}) could explain its lower overall GWP, despite energy production has been reported as the highest environmental impact related to the EASRPs. Vocciante et al (Vocciante et al., 2016; Vocciante et al., 2021a; Vocciante et al., 2021b) reported that almost the 73-76 % of the total GWP of an EASRP is related to the energy consumption. Conversely, the EASRP powered by the grid showed that only a 34 % of the total GWP was associated with the energy production. The mild powering in this case of study sheds light on a more sustainable remediation despite the use of an energy mix mainly made up of fossil fuels, as the Spanish grid, that has widely reported as potentially hazardous. Conversely, the intermittent powering of renewable energies reduces, stops or even reverses the electrokinetic mechanisms responsible of the pollutant transport from the soil to the wells, dropping the overall performance of the EASRP. The low efficiency of this intermittent powering makes necessary a higher power consumption and longer treatment times to transport the same amount of pollutant to the wells, reducing noticeable the sustainability of the process even though using a renewable energy as power source. Consequently, contrary to the promising results exposed by LCA analyses of electro-oxidation treatments coupled with green energies (Fernández-Marchante et al., 2021), those results claim against the use of renewable sources to power EASRP not only in terms of removal efficiency but also in terms of GWP.

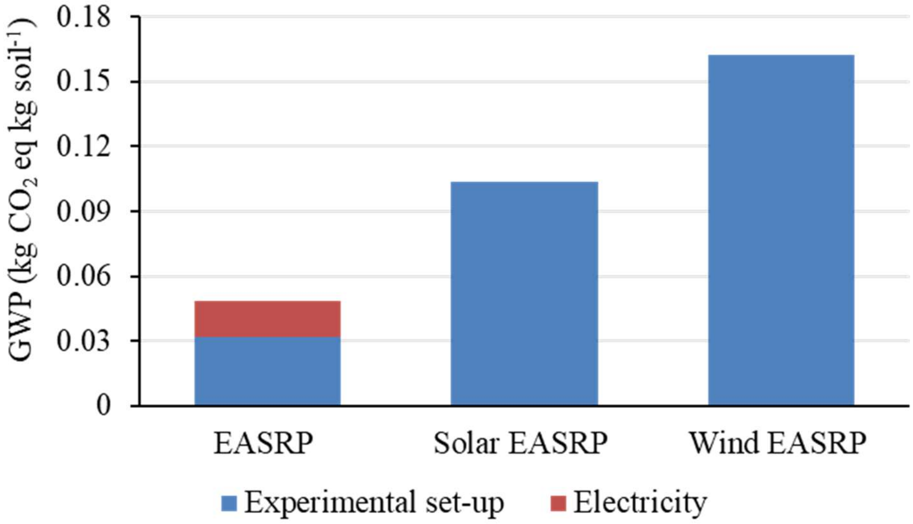

Figure 3. Global warming potential for an electrokinetic treatment under different powering strategies. Method: IPCC.

It is important to highlight that the energy and time required to remediate a polluted soil depends on the nature and properties of the soil and the pollutant (López-Vizcaíno et al., 2011). Hence the complexity and variability of a remediation treatment can bring out different environmental impacts according to those facts. Consequently, the GWP must be assessed in detail for each specific EASRP according to the soil and pollutant properties. In addition, as it was previously detailed, it is key to select the best functional unit that gives the most confident results.

Among the wide variety of gashouse emissions, halogen source gases which include chlorine and bromine contribute to ozone depletion due to their longer lifetimes in the atmosphere (Vallero, 2019). Those gasses destroy the ozone layer that preserves the earth of the UV radiation and prevents a temperature raise (Wexler, 2014). Consequently, to quantify the chlorofluorocarbons (CFCs) emissions is key to evaluate the environmental impacts of this process in the ozone layer and consequently to the global warming. Figure 4 shows the ozone depletion in terms of kg of CFCs per kg of soil treated.

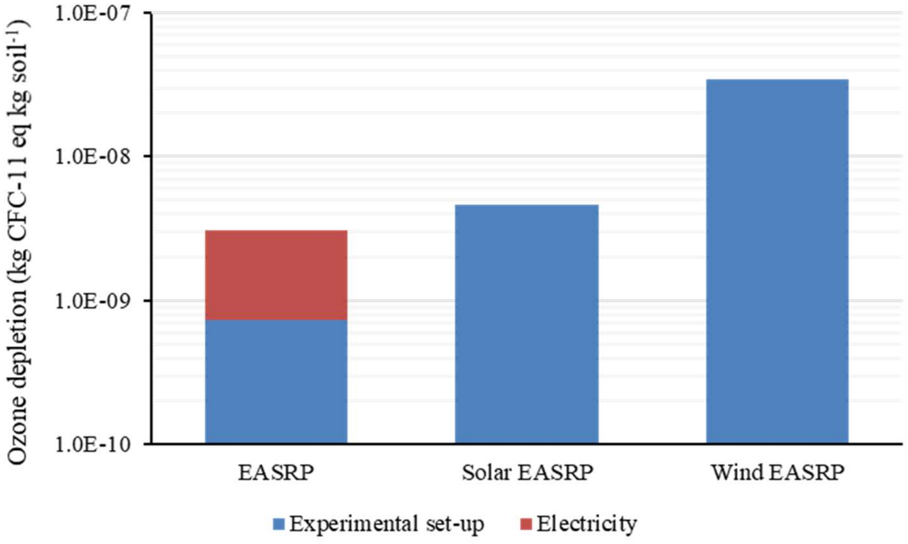

Figure 4. Ozone depletion in terms of kg CFC-11 per kg of soil. Method: ReCiPe.

As expected, the ozone depletion impact follows the same trend that the global warming potential. The massive emissions observed by the EASRP powered by the wind turbine turned into a higher ozone depletion, 34.2 µg of CFC-11 per kg of soil. Considering this value, a 13.6 % and a 9.0 % of the total ozone depletion showed by the EASRP powered by wind energy were observed by the EASRP powered by a PV plant and by the grid, respectively. According to the ozone depletion of the standard EASRP, a 76.3 % of the total ozone depletion impact is related to the energy production.

An additional parameter to be considered before making any statement regarding the sustainability of a process, is the water footprint. The research community has alerted to the water scarcity in the world, which can become into a humankind risk in the near future.

The United Nations estimates that by 2050 almost 1.8 billion of people may life under water scarcity and two-third of the world's population may hurt water stress conditions (United Nations, 2020b). Thus, it is essential to evaluate the water consumption related to the value chain of a process, product or service to reduce or get rid of it. Figure 5 shows the water consumption of the EASRP according to each powering strategy. Results show the highest water consumption for the EASRP powered by a wind turbine. In this case, 47.3 L kg soil^{-1} of water were required to achieve a remediation of 80.9 mg of 2,4-D per kg of dry soil. Concerning the EASRP coupled to the grid, data noticed that a 94.5 % of the total water consumption is related to the energy production, being almost negligible the water use related to the EASRP setup, 1.8 % of the total water use. Conversely, 21.3 L kg soil^{-1} and a 44.7 L kg soil^{-1} of water were used to remove 110.4 and 135.3 mg of 2,4-D per kg of dry soil using a PV plant as power supply and by means of the grid connection, respectively. As the previous results exposed, the use of a wind turbine showed the highest environmental impacts. The EASRP powered by PV panels or wind turbines shed light on huge environmental impacts purely due to the manufacturing of the energy production devices. Due to the huge influence of the energy production on the water footprint it is interesting to quantify the water consumption related to produce a unit of energy by the three power sources evaluated in this work. Thus, results state that 28.9 L of water are required to produce a kW energy considering a Spanish energy mix. On the other hand, 2.7 L and y 3 L are consumed to produce 1kW of energy using solar and wind power, respectively. Those results confirm that considering different functional units the results obtained in a LCA analysis could be completely different.

On the other hand, the lowest water consumption was observed by the PV powering, showing promising results in terms of water footprint. According to the water use required to perform the remediation treatment, as the electroosmotic flows noticed, the highest consumption of flushing effluent was reported by the EASRP coupled to the grid, 0.77 L of water kg^{-1} soil were transported during the 15 days of treatment. Thanks to the huge electroosmotic flow noticed during this EASR treatment, a higher pollutant dragging was carried out, reaching a huge level of remediation. Those data confirm once again that the intermittent powering of green sources leads to slowly and less efficient treatments which involve higher environmental requirements (water and energy) and consequently reports a lower sustainability. In line with the water consumption, the environmental toxicity of this pollution will differ depending on the powering mode used because of the pollutant dragging is mainly due to electroosmosis flows.

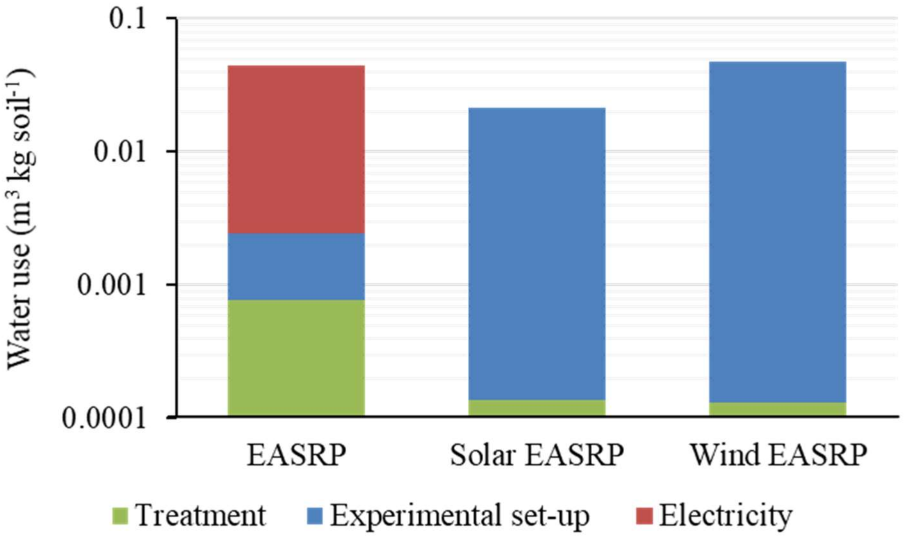

Figure 5. Water consumption related to each powering strategy. Method: AWARE.

In turn, it is important to evaluate the human toxicity of the pollution and the remediation treatments with the aim of assessing their environmental risks. It would not make sense to perform a remediation treatment that involves a higher environmental impact that the initial pollution. To test this possible issue, the toxicities of the initial pollution and the three proposed EASRPs were assessed. Figure 6 shows the non-cancer human toxicity after 15 days of treatment according to the different powering strategies studied and for the non-recovered pesticide. It is important to highlight that the toxicity analyses were assessed before and after 15 days of electrokinetic treatment. Thus, the remained pollutant concentrations into the soil matrix were different in each case of study. Results show that the toxicity of the polluted soil has a human impact of $4.83 \cdot 10^{-11}$ cases per kg of soil and this value is significantly reduced after the remediation treatment. It is important to note that the use of a wind turbine to power an EASRP only reduces the initial toxicity a 15.5 %. Nevertheless, working under a direct targeted powering mode for 15 days exposed a total toxicity reduction of a 39 % regarding the initial pollution. Despite the toxicity related to the pesticide dropped a 90 % in this case, the huge toxicity associated with the energy consumption (82 % of the total) increases the overall toxicity of the EASRP, up to $2.93 \cdot 10^{-11}$ cases per kg of soil. Those data claim that performing soil remediation treatments, regardless of the powering mode, reduces the human toxicity impact regarding the initial toxicity of the polluted soil. Nevertheless, the toxicity associated with the electricity generation and the experimental set-ups makes impossible to get rid of the initial pollutant toxicity without bringing out any additional environmental impact. However, in view of the results exposed, the use of solar power seems to be the best alternative to treat polluted soils in terms of human toxicity. Furthermore, longer treatment times, until a complete soil remediation, could shed light on a higher sustainability in terms of toxicity.

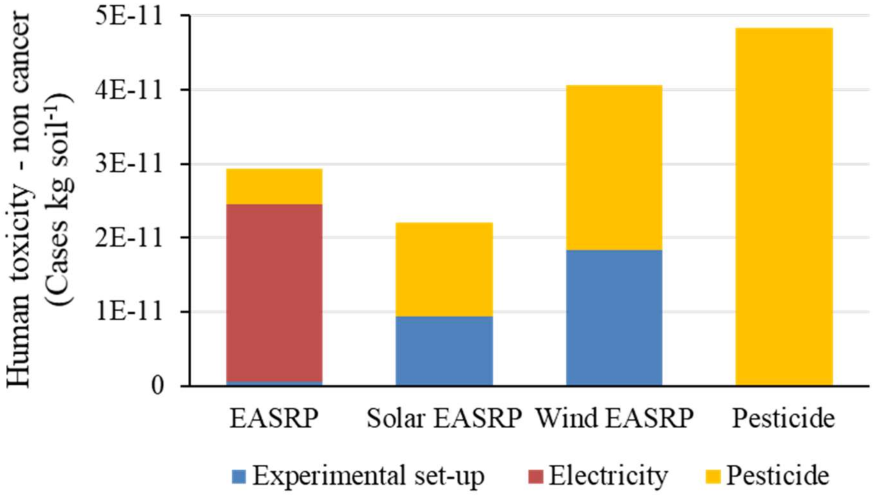

Figure 6. Human toxicity (non-cancer). Method: USEtox.

Apart from assessing the human toxicity, it is important to quantify the ecotoxicity of this pollution and its treatment in the freshwater. As aforementioned, the water scarcity is one of the most important environmental problems that the society must face up. This coupled with the fact that the pollution of water effluents or reservoirs can turn into an extremely dangerous environmental and health issue, to quantify its risks is key to control or stop their environmental impacts.

Considering this fact, the freshwater ecotoxicity was estimated as the potentially affected fraction of species (PAF). This impact category quantifies the fraction of species in an ecosystem that can be potentially affected by a high level of environmental pollution. It is important to note that a soil pollution can become into a groundwater pollution due to runoff and leaching processes. According to those facts, it is essential to evaluate the freshwater risk associate with this pollution. Results show that the freshwater ecotoxicity of the polluted soil is 0.0013 PAF m^3 day kg^{-1} soil. After 15 days of EASRP, the freshwater ecotoxicity related to the pesticide drops regardless of the powering mode, being noticeable by the grid powered EASRP due to the highest remediation reached in this case. On the downside, the effect of powering an EASRP with a wind turbine showed an increase of freshwater ecotoxicity, being 2.8 % higher than the initial pollution. Conversely, powering the EASRP by PV panels or by the grid tuned into a total ecotoxicity reduction of 51% and 43 %, respectively, regarding the initial pollution. In those cases, the freshwater ecotoxicity related to the pesticide dropped a 74 and 90 %, respectively. It is important to consider that almost 79 % of the total ecotoxicity of the grid-powered EASRP ($7.34 \cdot 10^{-4}$ PAF m^3 day kg $soil^{-1}$) is related to the energy production.

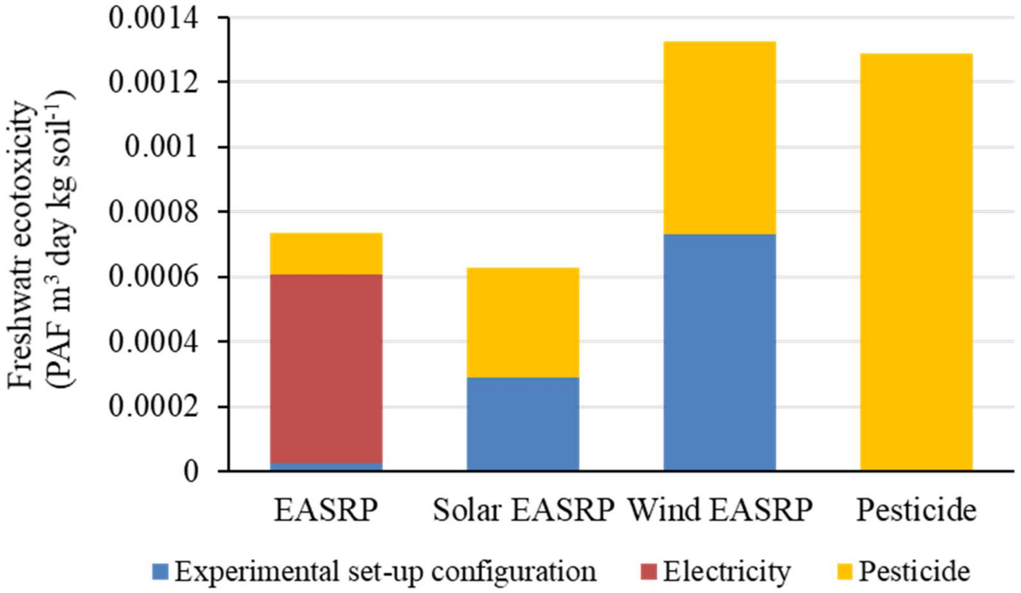

Figure 7. Freshwater ecotoxicity. Method: USEtox.

According to the freshwater ecotoxicity, the best strategy to power an EASRP is the use of PV panels, despite the huge energy requirements due to the reversibility of transport processes overnight, when no sunlight is available. It must be pointed out that increasing the treatment time could lead to a total soil remediation, dropping the ecotoxicity of the treatment until almost neglected values regarding the standard EASRP directly powered by the grid.

Finally, for a comparative propose, 17 endpoint impact categories were assessed by means of the ReCiPe method. Figure 8 shows the impact scores related to each endpoint impact assessed for the three powering strategies studied. Turconi et al. reported that the emission related to the production of energy may be related to different steps of the process depending on the nature of the power source (Turconi et al., 2013). The prominent impact of fossil fuel sources comes from the plant operation. On the other hand, the environmental impacts of renewable energies are strongly influenced by the setup manufacturing. According to these premisses and in view of the obtained results, the studied impact categories follow the same trend.

Results show that powering the remediation treatment using PV panels involves lower environmental risks. Despite the remediation reached under this powering strategy was not complete, longer treatment times could lead to a total soil remediation. Furthermore, the coupling of energy storage systems could smooth and ensure the EASRP powering throughout the day turning into promising remediation efficiency and lower environmental impacts.

According to the grid powered EASRP, it is important to highlight that despite the LCA was evaluated using a Spanish grid mix, which is mainly made up of fossil fuel (59.46 %), this powering strategy showed an environmental sustainability between the PV and wind powering. The low risks reported by this treatment could be directly associated with the lowest energy consumption required to reach a high remediation level by the same period of time. Nonetheless, keeping in mind the main environmental target of the EU, zero net emission by 2050 by means of a CO_2 free energy system, the impact related to the conventional EASRP treatment could be drastically reduced if the energy mix of the countries is almost made up of renewable sources.

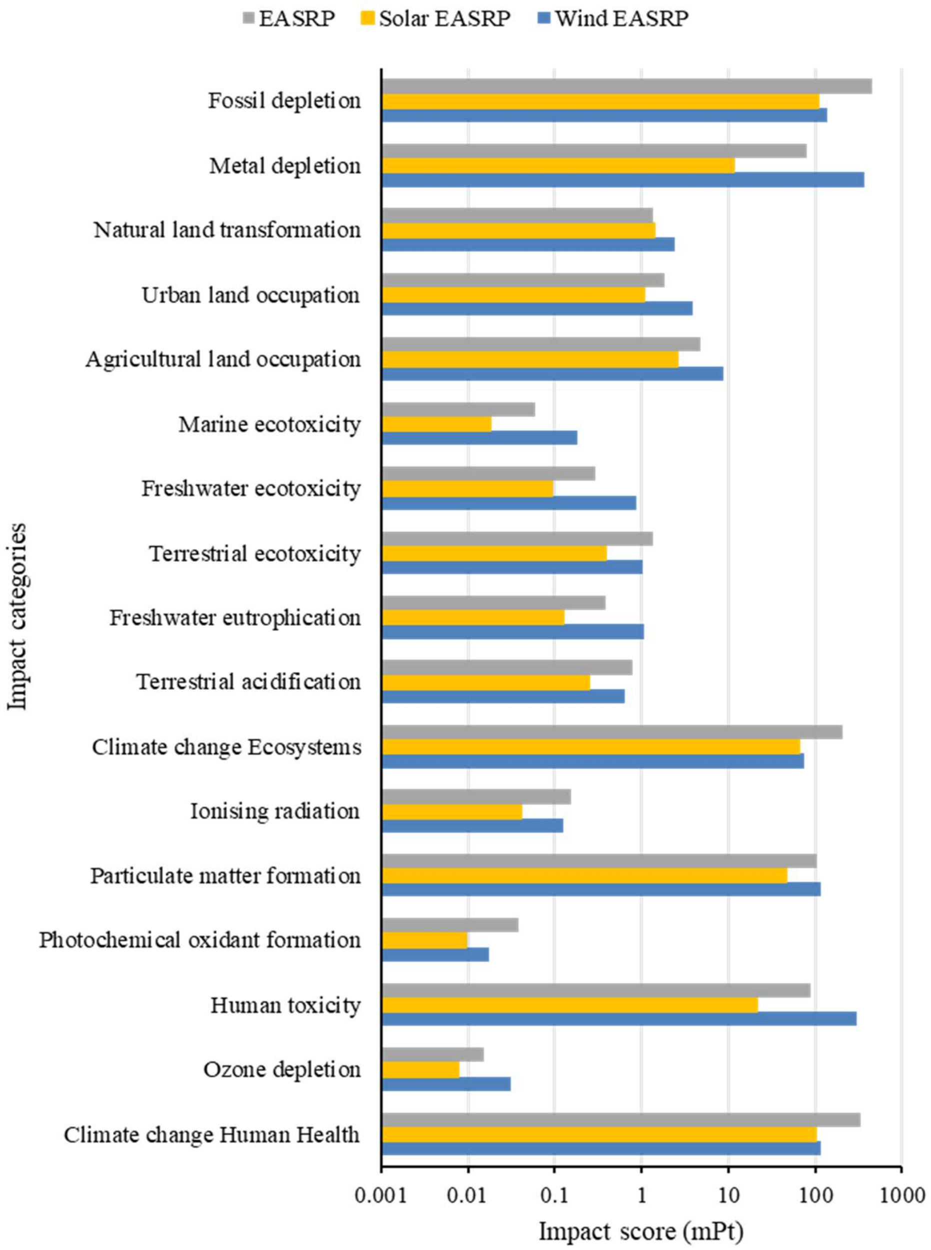

Figure 8. Effect of the powering mode on 17 endpoint impact categories. Method: ReCiPe.

In view of the overall endpoints assessed, results showed noticeable impact scores by the fossil and metal depletion and the human toxicity. According to the resource's depletion, fossil and metal, those impacts showed a converse trend. The environmental impact related to the fossil depletion is higher for the standard EASRP treatment which could be explained by the energy production according to a Spanish grid mix. Conversely, the metal depletion took a higher score for the wind powered EASRP which could be mainly due to the huge amounts of copper, iron and steel required to manufacture a wind turbine (Zhong et al., 2011).

On the other hand, climate changes impact categories, ecosystems and human health also showed meaningful impacts, being in both cases noticeable higher by the EASRP coupled to the grid. The energy production by a traditional energetic system based on fossil fuel sheds light on huge environmental and human risks which increases noticeable the impacts of those long-term categories. The rest of the studied environmental impacts noticed the same score trend, Wind >grid> PV. In order to reduce the environmental impacts related to the green energy production, new technologies must be researched. Furthermore, novel environmentally friendly material capable of being recycled at the end of its lifespan must be used to reduce the environmental risks associate with these technologies. In addition, it must be stated that the size of the green energy production setup has to be optimized according to the treatment that is going to be powered with the aim of taking advantage of the total energy produced, avoiding its waste and the drop of the performance and sustainability of the system.

Moreover, an optimization study of the energy supplied the EASRP was carried out and the environmental impact was assessed. Thus, the energy supply kept constant during the 15 days of treatment based on the good results obtained when grid energy was used, a treatment charge was 4.3 Ah kg^{-1}. In this way, the same efficiency and therefore the same removal of pesticides from the soil could be guaranteed. A lithium battery was added to the initial inventory (data obtained from database of SimaPro 9.0) to ensure a constant supply of energy during night-time or shutdown hours for the wind turbines. The energy density of the lithium battery is 37.9 Wh/kg (Weber et al., 2008). The solar panels as well as the wind turbines in the study were oversized in the experiments conducted, so this study only took into account the impact of these devices proportionally to the energy needed in the experiments (4.3 Ah kg^{-1}) allowing the remaining energy to be used for other purposes. Pessimistic scenarios for the application of renewable energies were chosen, taking into account information from the most climatically unfavorable regions. The study was carried out considering an average power of the solar panels of 100W and the wind turbine of 200W when then can achieve 300W and 600W, respectively. This energy optimization and regulation allow to obtain better results also from the point of view of overall environmental impacts, as can be seen in the following Table 2.

Table 2. Uncertainty analysis of the midpoint results. Values are presented per kg of soil

		EASRP Spain	**Solar/ battery EASRP**	**Wind/battery EASRP**
Climate change	kg CO_2 eq	48.7	47	46.7
Ozone depletion	µg CFC-11 eq	3.08	2.52	2.71
Water use	m^3	45	42.4	42.4
Human toxicity, non-cancer	cases	2.93E-11	2.96E-11	3.03E-11
Freshwater ecotoxicity	PAF.m^3.day	0.000734	0.000770	0.000771

Thus, it can be concluded that despite the use of PV panels to power an EASRP shows a higher GWP, the rest of environmental risks noticed the lowest impact by the use of this renewable energy to power an electrochemically-assisted soil remediation process. However, if the design of the renewable devices is suited to the energy needs of the treatment and an energy storage system is installed, the impact of the carbon footprint is also reduced. Consequently, this powering strategy could become into a promising alternative to perform in-situ EASRPs in places where there is not access to the grid system once the PV plant is designed according to the electrical requirements of the electrochemical treatment.

Given the wide variability of goals and functional units that can be studied, comparing those results to other reported in literature is highly complex. Nevertheless, several research groups have evaluated the environmental impacts related to soil remediation treatments. Trentin et al. (da S Trentin et al., 2019) assessed the impacts of phytoremediation, excavation and electrokinetic treatments under the three pillars of sustainability (environmental, economic and social). Results noticed the lowest environmental and economic impacts to the phytoremediation technique. Nevertheless, the longer times required to perform a complete recovery of the soil under this technology reduce its social sustainability. Conversely, this sustainable pillar plays an important role in the EASRP due to its faster removal of pollutant. In addition, it is worth mentioning that one of the main advantages of a EASRP is that it can be carried out in-situ. This operational strategy reduces the impacts of any technology regarding an ex-situ treatment that sometimes may affect the human health and ecosystem quality more than not taking remediation actions (Mauko Pranjić et al., 2018).

As aforementioned, the electrochemical remediation depends on the pollutant and soils properties. For this reason, a complete analysis (technique and environmental) must be performed before selecting the most suitable remediation technology to recover a particular polluted soil.

Conclusions

The LCA analyses of grid and green powered EASRPs have shown interesting conclusions in terms of sustainability. Data notice lower GWP values by the traditional grid powering (49 g of CO_2 eq. per kg of soil), being a 34 % of the total emissions related to the energy production. These surprising results can be explained by the high amount of energy supplied to the wind powered EASRP during the 15 days of treatment, 10 times higher than by the grid powering mode. The direct connection of a green energy production device without a detailed design according to the treatment that is going to be powered, may lead to an uncontrolled remediation process because the energy supplied has been misused. Keeping in mind those facts, the high CO_2 emissions of the wind powered treatment could be explained.

Regarding the water consumption associated with each powering strategy, results showed the lowest water consumption by the solar powered EASRP (21.3 L kg soil^{-1}). In addition, it is important to highlight that a 94 % of the total water consumption of the grid powered EASRP, is related to the energy production. Furthermore, it worth mentioning that the water consumption associated with the flushing effluent was in line with the electro-osmotic flows and the level of remediation reached by each treatment. On the other hand, toxicity estimations pointed out that between a 54 and 90 % of the initial toxicity of the pollutant was removed from the soil, being higher by the grid powered EASRP. Nevertheless, the huge toxicity related to the setup manufacturing and the energy production increases the overall toxicity of the process.

According to the main conclusions drawn in this study it worth noting that the environmental impacts of a process have to be assessed once the treatment is fully optimized. Furthermore, reliable results may only be obtained considering the most accurate functional unit that allows to compare technologies to reach confident conclusions.

Acknowledgements

Financial support from the Spanish Agencia Estatal de Investigación through project PID2019-107271RB-I00 (AEI/FEDER, UE) is gratefully acknowledged. M. Millán thanks the UCLM for the pre-doctoral contract within the framework of the Plan Propio I+D.

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