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Comparative environmental and economic Life Cycle Assessment of phytoremediation of dredged sediment using *Arundo Donax*, integrated with biomass to bioenergy valorization chain

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Abstract

The economic and environmental life cycle assessment (LCA) was integrated into a laboratory-based experiment to evaluate the feasibility and sustainability of phytoremediation of chloride-rich marine dredged sediment, using perennial reed *Arundo Donax* along with biomass valorization. As a prerequisite for life cycle assessments, a baseline mathematical model was developed to estimate the yields of biomass to bioenergy valorization chain including the estimation of biomass yield per m³ sediment, bioenergy yields from valorization schemes, expected green electricity yield, and the phytoremediation time frame. This mathematical model was applied to develop a parametric life cycle inventory for two scenarios of sediment phytoremediation separately or integrated with biomass valorization, for LCA and further sensitivity and uncertainty analysis. Comparative LCA unveiled that the cost and environmental impacts of annual phytoremediation of 1m³ sediment alone or integrated with biomass valorization are much inferior to the corresponding sediment landfill as the inevitable alternative approach for sediment management. With the chloride bioaccumulation capacity of 9940 mg per kg dry biomass of *A. donax*, the phytoremediation

of sediment with chloride concentration higher than 1650 mg/kg may not be achievable in a realistic time frame. Due to the importance of considering sediment depth and the effectiveness of the plant rooting system in estimating the performance of phytoremediation and the time frame, the volume of sediment (1m^3) is a more appropriate functional unit than the surface area (ha) for LCA studies of phytoremediation. In addition, considering the volume of sediment as a functional unit retains comparability to other valorization scenarios such as sediment incorporation in cementitious matrices and management scenarios such as landfill, which are generally expressed on a volume or mass basis. Integrating biomass-derived bioenergy production into phytoremediation could offer local and global benefits in terms of economy and environment mainly due to carbon sequestration and avoiding fossil-based fuels.

Keywords: *Arundo donax*, life cycle assessment, chloride-rich marine dredged sediment, Biomass to bioenergy valorization, phytoremediation

1. Introduction

As a worldwide issue in terms of economy and environment, the million tons of annually dredged sediments must be managed to satisfy the gradually stricter regulatory requirements. According to French regulations, the discharge of polluted sediments and dredging residues into the sea would be prohibited from 2025 (legifrance, 2016). The non-inert dredged sediment carried to land should be treated via remediation, valorization, landfill, or combined approaches. Along with the conventional physicochemical remediation technologies which are very costly for sediment with a zero-market value and may not be feasible on a large scale, the passive treatment of phytoremediation has been identified as a cost-effective, eco-friendly, solar-driven and in-situ technology for bio-remediation of contaminated sediment

(Zaghloul, 2020). The process consists of using plants and their associated micro-organisms to reduce contamination from a given media by uptake and bioaccumulation (Pirzadah et al., 2015). The long-term time frame and lack of revenue from the land during the remediation process are the major bottlenecks of phytoremediation for further popularity and development. To alleviate these limitations, phytomanagement emerged from integrating biomass valorization into phytoremediation (Nascimento et al., 2021). Phytomanagement of dredged sediment, under the concepts of circular economy, could synergize biomass production and carbon sequestration with phytoremediation, offering social, economic, and environmental benefits during the whole remediation process up to inert and productive sediment (Nascimento et al., 2021).

Two general types of hyperaccumulator and high-yielding biomass plants could be applied for in-situ phytoremediation of dredged sediment to reduce the targeted contaminants to regulatory levels. The proper plant species for sediment phytoremediation could be selected from a compromise between the plant accumulation rate of target element, growth rate, biomass production capacity, biodiversity, tolerance to high concentrations of accumulated elements and salinity of sediment (Rabêlo et al., 2021). Since dredged sediments are generally multi-element contaminated, the hyperaccumulator plants may not be tolerant to multiple inorganic compounds such as heavy metals (Nascimento et al., 2021). *Arundo Donax* (*A. donax*), a high-yielding perennial reed, highly efficient resource utilization, tolerant to biotic, abiotic and salinity stresses, trace elements and chemical composition of sediment has been applied for phytoextraction of pollutants along with the production of marketable biomass (Bosco et al., 2016). Due to the growth capability in contaminated and infertile soils and the potential of heavy metals bioaccumulation (Cristaldi et al., 2020), *A. donax* has been promising for phytoextraction of contaminants from soil such as Hg (Sagehashi et al., 2011; Corno et al., 2014), Cd (Yang et al., 2012; Sabeen et al., 2013; Cristaldi et al., 2020), Cr

(Kausar et al., 2012), As (Yang et al., 2012), Pb (Guo and Miao, 2010; Yang et al., 2012), Ni (Papazoglou et al., 2007; Atma et al., 2017), Mn, Zn and Fe (Corno et al., 2014a). The spontaneous natural growth of *A. donax* in the saline dredged sediments witnessed in the south of France (Fig. S1) was apparent evidence of the capability of *A. donax* to adapt to chloride-rich and polluted sediment as a medium for growth. However, the feasibility of chloride phytoremediation of dredged sediment by *A. donax* as well as its bioaccumulation capacity is little known to be nominated as a chloride phytoremediation plant (Calheiros et al., 2012; Jesus et al., 2017; Andreu-Rodríguez et al., 2017). Recently, a laboratory-based experiment has been implemented to address the growth feasibility of *A. donax* in marine dredged sediment and to quantify the bioaccumulation rate of chloride in the leaves and steams of *A. donax* (Sordes et al., 2021). In this study, beyond feasibility, the sustainability of chloride phytoremediation of sediment by *A. donax*, alone and combined with biomass valorization was aimed to be assessed in terms of economy and environment through a comparative life cycle assessment (LCA). The LCA has been applied in some studies as a common approach to assessing the environmental impacts of phytoremediation (Nie et al., 2010; Suer and Andersson-Sköld, 2011; Witters et al., 2012; Vigil et al., 2015; Mattei et al., 2018; O'Connor et al., 2019). However, LCA of phytoremediation of chloride-rich sediment, as a common pollutant in all marine dredged sediments is still lacking. The economic and environmental assessments of phytoremediation as well as biomass to bioenergy valorization chain, combined with laboratory-based phytoremediation trials, offer a holistic approach to study the feasibility, sustainability, the performance of sediment phytoremediation.

This study presents a part of a comprehensive ongoing project for the port of Camargue in the region of Occitanie in the south of France to provide insight into the feasible and sustainable remediation and valorization strategies for marine dredged sediment. As a prerequisite for economic and environmental assessments, a mathematical model was developed to estimate

the time interval for phytoremediation of multiple pollutants as well as the yield of biomass to bioenergy valorization chain including the estimation of biomass yield, bioenergy yield from valorization schemes and green electricity yield per m³ of sediment. The mathematical model was applied to develop life cycle inventories (LCIs) for the chloride phytoremediation of marine dredged sediment. The objective of this study was to integrate the economic and environmental LCAs into the laboratory trials to offer a comprehensive insight into the environmental sustainability, economic viability, and technical feasibility of sediment phytoremediation by *A. donax*. separately or combined with biomass valorization. This integrated approach could be customized for other plant species, pollution types, as well as biomass valorization scenarios.

2. Material and method

2.1 Biomass valorization to bioenergy

To fulfil the requirements of developing life cycle inventories for harmonized economic and environmental life cycle assessments, adapted to the functional unit of 1m³ sediment phytoremediation, a mathematical model was developed to estimate biomass yield, bioenergy yield, green electricity generation as well as estimation of phytoremediation time frame (equation of 1 to 8).

2.1.1 Biomass yield

The quantity of dry biomass per one cubic meter of sediment (kg/m³) per harvest could be calculated from equation 1.

$$dm = \left(\frac{PD * (SR/100) * FB * (1 - MP/100)}{H} \right) \quad (1)$$

In which **dm** is the average yield (kg/m³) of dry mass expecting from each m³ of sediment per harvest, **PD** is the density of plant cultivation per m², **SR** is the survival rate (%) of the plants, **FB** is the average quantity (kg) of fresh biomass per plant per harvest, **MP** is the percentage of the moisture content (%) in fresh biomass, **H** is the depth (m) of sediment (soil).

The range of plant density of 0.5 to 1 for temperate and 1 to 2 plants per m² for warmer climates (5000-20000 per hectare) are reported (Ge et al., 2016; Corno et al., 2014). The planting depth for *A. donax* is recommended to be between 0.15 m to 0.25 m (Corno et al., 2014). The moisture content of fresh biomass of *A. donax* is reported to be between 35 to 52 percent by weight (Ragaglini et al., 2014; Ge et al., 2016) with an average percentage moisture (MP) content of 49.2 percent on a mass basis (Melane et al., 2017). The yield range of 15 to 41ton dry mass per hectare for fall harvest and 21 to 49ton dry mass for winter harvest is reported (Nassi O. Di Nasso et al., 2011; Ping et al., 2014; Ge et al., 2016).

In this study, the biomass yield of 20 tons of dry mass per hectare per year, a surface plant density of 1 plant per m² and a sediment depth of 0.2 m are adopted based on the literature. *A. donax* is highly resistant to salinity and the toxicity symptoms appear only after severe salt stress with zero plant mortality (De Stefano et al., 2018). In addition, *A. donax* has the capability to cope with a wide range of multiple soil and environmental stresses and fast recovery following stress relief (Cocozza et al., 2020). Nevertheless, the survival rate (SR) of the plant in the extreme condition of multiple stress is incorporated as a parameter into the model with the initial value of 97 percent (Zhang et al., 2021).

2.1.2 Duration of phytoremediation

The maximum time frame required for remediation of sediment with q pollutant elements up to expected concentration thresholds could be estimated from equation 2.

$$y = \max_{1 \leq i \leq q} \left\{ \frac{\rho_s (p_{ec_i} - p_{er_i})}{dm * f * d_r} \right\} \quad (2)$$

In which y is the maximum years expected for the phytoremediation, ρ_s is the density (kg/m^3) of sediment (soil), p_{ec_i} is the current concentration ($\text{mg/kg}_{\text{sediment}}$) of i^{th} pollutant element in sediment, p_{er_i} is the regulatory critical concentration ($\text{mg/kg}_{\text{sediment}}$) of i^{th} pollutant element in sediment in $\text{mg/kg}_{\text{sediment}}$, dm is the average yield (kg/m^3) of dry mass expecting from each m^3 of sediment per harvest, f is the frequency of harvest per year, d_r is the deposition (bioaccumulation) rate ($\text{mg/kg}_{\text{dry biomass}}$) of the i^{th} pollutant element in dry biomass of the plant, q is the number of pollutant elements in the sediment.

The density of sediment (ρ_s) in this study is considered to be 2270 kg/m^3 based on the laboratory sediment characterization. The current concentration of leachable chloride (Cl^-) measured after partial dewatering in geotube was $16000 \text{ mg per kg sediment}$. According to French regulations, the concentration threshold of leachable chloride in inert sediment is 800 mg/kg (legifrance, 2016; Delattre et al., 2022). The frequency of harvest per year (f) is reported to be 1-2 times per year (Lewandowski et al., 2003; Ge et al., 2016), two harvests per year ($f=2$) were adopted in this study.

2.1.3 Biomass valorization to bioenergy

The bioconversion industry has been developed in four generations to transform biomass into clean and affordable biofuels or chemicals (Demirbas, 2011). First generation biofuels are synthesized from edible biomass cultivated in arable land with the potential pressure on food

security. To mitigate the environmental impacts, the usage of arable land and food crops as the drawbacks of first-generation biofuels, second-generation biofuels are obtained from non-edible biomass such as perennial grass which could be cultivated in marginal land, as well as agricultural waste containing lignin and cellulose known as lignocellulosic biomass. The third and fourth generation biofuels are produced from algae (Sikarwar et al., 2017).

To compete with fossil fuels in terms of economy, low cost non-edible lignocellulosic biomass is a pre-requisite for biofuel production at a commercial scale. Perennial energy crops could have advantages over annual crops in terms of output to input efficiency of energy and cost thanks to the allocation of overhead expenses over plant exploitation (cultivation) lifetime (Angelini et al., 2005). Perennial grasses can easily sprout again after aerial part harvesting even when they are exposed to toxic trace elements, with no need for replanting (Rabêlo et al., 2021). Among perennial energy crops producing lignocellulosic biomass, adapted to the Mediterranean environment, the non-food energy crop of *A. donax* could be utilized as a second-generation energy feedstock for bioenergy production including biogas, bioethanol, combustible solid as well as the raw material for green chemistry industry (Corno, 2016).

The potential energy theoretically obtained from the biomass harvested from phytoremediation of 1m³ of sediment could be calculated from equation 3.

$$BE = dm * HCV * f \quad (3)$$

In which **BE** is the average of potential bioenergy from phytoremediation of one m³ of sediment per year, **dm** is the average yield (kg/m³) of dry mass expecting from each m³ of sediment per harvest, **HCV** is the heat calorific mean value (kJ/kg dry biomass), and **f** is the frequency of harvest per year.

The heat calorific mean value of *A. donax* is reported around 16.8 MJ per kg dry weight (Krička et al., 2017).

2.1.4 Bioenergy yield estimation (Biogas, biomethane, bioethanol and solid combustible yield)

Biomass, as the only renewable resource of carbon, can be transformed into solid, liquid, and gaseous fuels as alternative energy sources to fossil fuels, through thermochemical and biochemical pathways (Saikia et al., 2015). The bioenergy yield in the forms of gas, liquid, and solid biofuels from the biomass of *A. donax*, could be obtained from equations 4, 5, 6 and 7.

$$\text{Solid combustible yield (kg)} = DM \quad (4)$$

$$\text{Bioethanol yield (liter)} = DM * BETY \quad (5)$$

$$\text{Biomethane yield (Nm}^3\text{)} = VS * BMY \quad (6)$$

$$\text{Biogas yield (Nm}^3\text{)} = VS * BGY \quad (7)$$

In which DM is dry biomass (kg) of *A. donax*, BETY is bioethanol yield (litre) per kg dry biomass, VS is volatile solid (kg) of biomass, BMY is biomethane yield (Nm³) per kg volatile solid of *A. donax*, and BGY is biogas yield (Nm³) per kg volatile solid of *A. donax*.

Having approximately the same heat energy value as wood (18 MJ/kg), *A. donax* has a great potential for direct burning as a combustible solid fuel.

The specific biomethane yield (BMY) of 0.185 normal cubic meters (Nm³) (Corno, 2016), the BMY range of 0.151 - 0.391 Nm³ (Corno et al., 2014; Dragoni et al., 2015; Ge et al., 2016) and the highest biochemical methane potential of 0.392 Nm³ (Ragaglini et al., 2014) per kg of volatile solids, are reported from the anaerobic digestion of *A. donax* biomass. The volatile solids (VS) of biomass could be determined from total solid (TS) biomass obtained from thermal treatment of biomass at 105°C at a constant weight, minus ashes obtained from

thermal treatment at 550°C for 10 hours (Ceotto et al., 2021). Biogas yield (BGY) of 0.272 Nm³ and biomethane yield (BMY) of 0.174 Nm³ per kg of total solids (TS) are reported from anaerobic digestion of *A. donax* biomass (Corno et al., 2015). An estimation of approximately 0.075 liters or 0.06 kg of bioethanol yield per kg dry biomass (BETY) of *A. donax* is presented by Silva et al. 2015 (Lemons e Silva et al., 2015).

2.1.5 Green electricity yield estimation

The quantity of renewable electricity from valorization bioenergy schemes, based on kilowatt hour (kWh), could be obtained from equation 8:

$$\text{Green electricity yield (kWh)} = HCV * BEY * PPE / 100 * 1 / 3600 \quad (8)$$

In which **HCV** is heat calorific mean value on kJ/kg for combustible solid, kJ/l for bioethanol, kJ/Nm³ for biomethane and biogas, **BEY** is bioenergy yield in forms of solid (kg), liquid (l) or gas (Nm³), **PPE** is efficiency of bioenergy power plant in percentage (%). Since every hour is 3600 seconds, the quantity is divided into 3600 to convert kJ to kWh.

The efficiencies of power plants are diverse depending on the source of fuel and generator technology. In this study, the cogeneration of heat and power (CHP) plants was adopted to convert biogas to green electricity. CHP systems typically have total efficiencies between 65 to 80 percent and could be approaching 90 percent (European Commission energy). The bioenergy power plant in this study is considered to be a CHP power plant with the efficiency of 50 percent (Payet and Ribak, 2008).

2.2 Phytoremediation by *A. donax*

2.2.1 *Arundo donax*

A. donax or giant reed (giant cane) is a perennial grass with an outstanding growth rate and biomass yield and low requirements in terms of fertilizer, pesticide, irrigation and soil tillage, along with environmental plasticity to adapt to wide ranges of pH, salinity, drought and extreme growing conditions (Krička et al., 2017; Ceotto et al., 2021). In spite of having the C₃ photosynthetic pathway, *A. donax* exhibits an unusual high photosynthetic CO₂ sequestration rate and biomass production, higher than other C₃ grasses and similar to C₄ species (Sidella, 2013). Its growth rate reaches up to 7 cm per day in summer in Mediterranean environments (Bassam, 2010). Under natural wild conditions, *A. donax* would yield around 29 t/ha of dry biomass (Angelini et al., 2009; Bassam, 2010) which could reach up to 55 t/ha or the highest-ever yield of 125 t/ha (Idris et al., 2012) under cropping conditions (Ververis et al., 2004; Nassi o Di Nasso et al., 2010; Ceotto et al., 2021).

The industrial applications of the lignocellulosic biomass of *A. donax* as a feedstock for second-generation biorefineries such as biomethane, bioethanol and combustible solid (Zhang et al., 2021; Krička et al., 2017), as well as cellulose production for pulp, paper and biopolymer industry (Jámbor and Török, 2019;), has been proven. The cell wall polymers of lignin, cellulose and hemicellulose are the main three constituents of the complex matrix of lignocellulose biomass. The higher the lignin content, the more preferable for direct combustion, while the higher the rest means more suitable for gas and liquid fuel production (Krička et al., 2017).

Due to the high carbon and hydrogen contents, as the basic elements of any fuel, and the high carbon to nitrogen ratio, as well as the holocellulose to lignin ratio *A. donax* has been addressed as one of the best species for biogas production (Sánchez et al., 2021) comparable to conventional annual crops like maize (corn) (Gierl et al., 2022; Corno et al., 2014) and bioethanol production (Jámbor and Török, 2019). Moreover, the high calorific mean value of

the biomass, independent of the growth condition (Angelini et al., 2005) promises the potential of *A. donax* for direct combustion. The plant versatility, the diverse bio-refinery products, and the high yield of biomass as a carbon-neutral energy source contributing to the climate-neutrality (Verónica et al., 2022), nominated *A. donax* among the eligible candidates to be integrated into phytoremediation.

2.2.2 Experimental phytoremediation of marine dredged sediments by *A. donax*

To study the capability of *A. donax* in the in-situ chloride phytoremediation of marine dredged sediment, laboratory-based pot cultivation of *A. donax* was performed. The objective was to investigate the chloride bioaccumulation performance of *A. donax* in natural conditions and chemically assisted cropping conditions via adding mycorrhiza to the sediment as well as humic acid as an alternative to synthetic chelators. Due to not propagating by seeds, two asexual reproduction types of rooting at the nodes and the cane fragment were applied for the vegetative reproduction of *A. donax* (Salazar-Zeledón et al., 2015). The reproduction process was performed within a nursery, accompanied by the partner company of Nymphéa Distribution (nymphheadistrib.com). After 2 months of growing in peat in 3L pots, *A. donax* plants were acclimatized for two weeks in the greenhouse and were bathed before being transplanted into the 5 litres of dredged sediments. Dewatered dredged marine sediment from an ongoing pilot project in port Camargue was used as peat-free cultivation media for in-situ phytoremediation using *A. donax*.

The sediments dredged from port Camargue (Fig. S2) were sampled after dewatering in a geotube with the chloride concentration of 1.6% of the chemical composition and 33% water content. Crystalline powder of Humic acid (CAS: 1415-93-6. EINECS: 215-8096) from the company Alfa Aesar (alfa.com/fr/catalog/041747/) and Mycorrhiza (MYCOR®) from the company IfTech (iftech.fr/mycor-en-pot-de-23-l-c2x1779336) were supplied. *A. donax* plants

were introduced into 4 cultivation media distinguished by 1) sediment without any additive, 2) sediment with the addition of mycorrhiza, 3) sediment with the addition of humic acid, and 4) sediment with the addition of mycorrhiza and humic acid (Sordes et al., 2021). A plant repotted in the same supplier's peat mixture without any sediment was set up as a control. Pots were set up in quadruplicate as illustrated in Fig. 1.

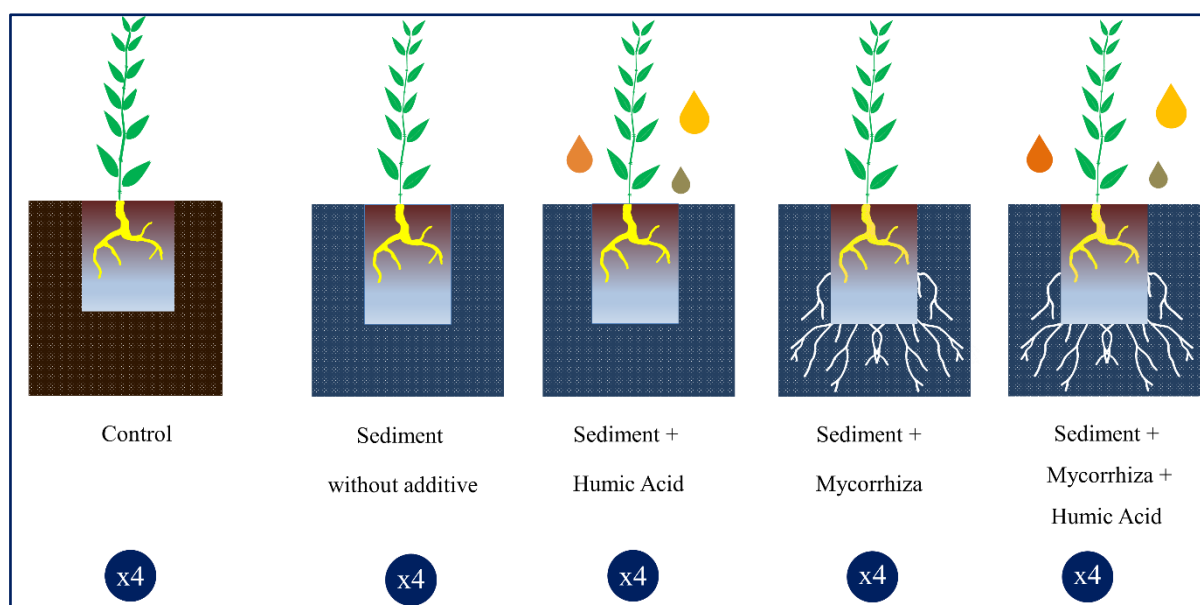


Fig. 1. Schematic view of *A. donax* cultivation in quadruplicate pots with distinct culture medium (Sordes et al. 2021)

At the end of the experiments, the *A. donax* plants were photographed, the aerial parts harvested, and treated for further chloride extraction and ion chromatography according to the protocol of McSorley 2016.

2.3 Environmental assessment

2.3.1 Life cycle assessment

Life cycle assessment (LCA) is the most popular and recognized analytical framework for comprehensively assessing the environmental impacts and to identify the hotspots associated with the entire life cycle of a product or process (Guinee, 2002; Soleimani et al., 2023). The comparative attributional life cycle assessment was carried out according to ISO 14040, 2006

and ISO 14044, 2006 in four standard stages of goal and scope definition, inventory analysis, impact assessment and interpretation. The life cycle inventories (LCIs) of the phytoremediation scenarios were developed in openLCA v. 1.11.0, based on laboratory and field data, the literature and by taking advantage of the inventories from the database ecoinvent v.3.5 with allocation at the point of substitution (APOS). The ReCiPe midpoint (H), a state-of-the-art method for life cycle multi-impact assessment harmonized at the midpoint and endpoint level (Huijbregts et al., 2017) was applied to assess environmental impacts comprises 18 categories at the midpoint level. At the endpoint level, the midpoint impacts are accumulated in three main damage categories of human health, resource, and ecosystem diversity. The environmental impacts of phytoremediation were applied for the normalization to convert the impact categories with different units into a unitless score by dividing each impact value to its corresponding of phytoremediation. In addition, the impacts of 1m³ of sediment landfill in France were normalized to the corresponding impacts of both phytoremediation scenarios. The average of normalized impacts is adopted as the single score environmental impact and illustrated in a graph to provide comparative insight into the difference in the order of magnitude between the impacts of the landfill and phytoremediation scenarios.

2.3.2 Goal and functional unit of the LCA

Goal and scope definitions give a descriptive big picture of the production system in terms of system boundary and functional unit. A functional unit quantifies the performance of a product system as a reference unit to ensure comparable LCA studies and reliable results. Accordingly, in-situ treatment of one cubic meter (1m³) of sediment was adopted as the functional unit in this study. All the flows coming into the system boundary such as energy, transport, water and raw materials and outgoing flows including emissions to air, soil and

water were considered. All the processes in the phytoremediation and biomass valorization scenarios were considered in the cradle-to-gate system boundary illustrated in Fig.1. The system boundary does not include the laboratory-based growing of plants and the associated trials. As a cut-off approach, the sediment was considered to be a recyclable material with zero environmental impacts, allocating all the environmental impacts up to the end of sediment dewatering in the geotube on land, to the dredging operation. To provide more insight into the sensitive parameters, the results of sensitivity analysis and uncertainty analysis are presented in this study. For the reliability of the comparative assessments between scenarios, consistency was sustained in all parameters and inventories.

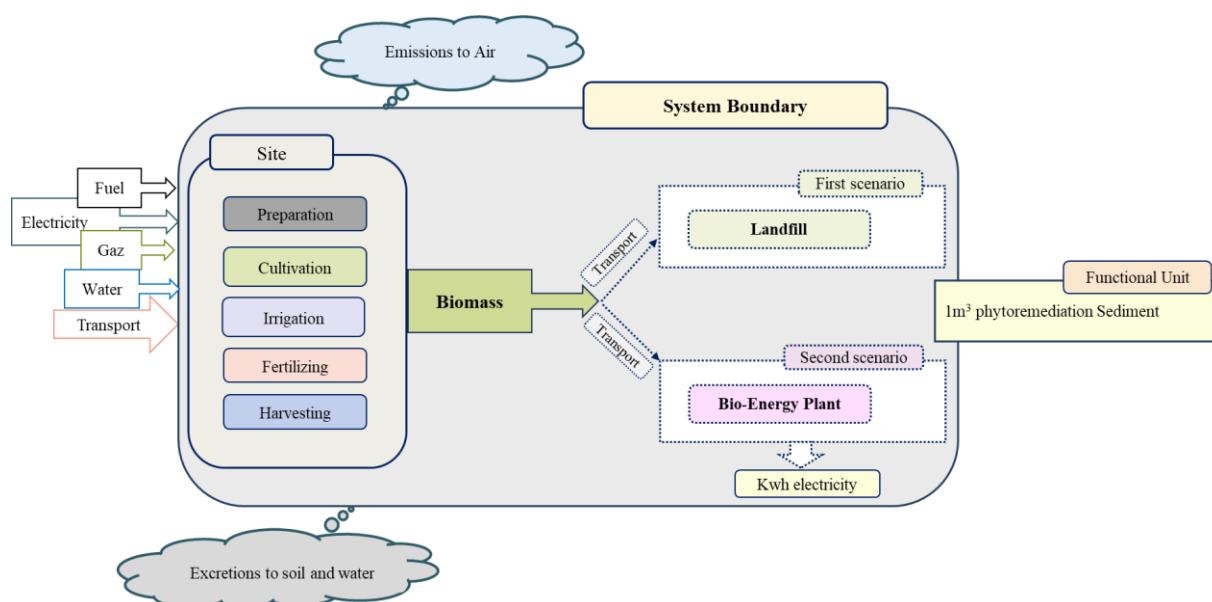


Fig. 2. Scheme of the system boundary, which includes the site preparation, production of plants, planting, watering, fertilizing, harvesting, input raw materials, bioenergy power plant, transport of materials to the power plant, and the green electricity production in the power plant.

Among the biomass to bioenergy valorization schemes the biogas production from biomass was adopted as a biomass valorization approach in this study. To incorporate the impacts of

biomass transformation to biogas and ultimate green electricity, it was assumed that the same quantity of electricity produced with the fossil fuel of natural gas in the same power plant is avoided. In this way, the economic and environmental benefits of integrating biomass valorization into phytoremediation arises from the replacing of fossil fuel (natural gas) with synthetic biogas for green electricity production in power plant.

2.3.3 Data (biomass characterization and bioenergy production)

The physicochemical and geotechnical specifications of the sediment samples of the port Camargue were characterized by the CISMA environment laboratory in France (Table S1). The analysis confirmed that sediments are non-hazardous but not inert in terms of some pollution elements including chloride. The mean values of the materials and energy flows and the processes within the system boundary were adopted from the project internal data, laboratory analysis as well as background data. The transport of raw materials and harvested biomass were estimated from the origin to the destination using Google Maps.

The agricultural activities such as soil preparation, cultivation, fertilization, harvesting, machine operations and transport to the anaerobic digester biogas power plant of EveRé (evere.fr), 97.5 km from the port of Camargue are included in the assessment. The chloride-rich biomass is transformed into synthetic biogas through an anaerobic digestion plant and the biogas was assumed to be burnt in a combined heat and power (CHP) plant also known as cogeneration to produce electricity. In the scenario of phytoremediation, it is assumed that the harvested biomass would be landfilled in a site (Bellegarde) with 53 km distance from the port of Camargue. The volume quantity of biogas per kg dry mass of *A. donax* was estimated based on 0.272 Nm³ biogas per kg total solid (Corno, 2016). The annual biomass yield was assumed to be 20 tons of dry mass per hectare from two harvests per year.

2.4 Uncertainty Analysis

The uncertainty in the parameters may result in overestimation or underestimation of the environmental impacts. A Monte Carlo (MC) simulation is a tool to obtain the probability distribution for each environmental impact category due to the combined uncertainties of the model parameters. Random sampling from the probability distribution functions of the incorporated parameters in the model, MC could comprehensively assess the sensitivity of the environmental impact categories to the uncertainties in the life cycle inventories (Soleimani et al., 2023). Monte Carlo simulations in openLCA unveiled the confidence interval in each impact category. The 90% confidence interval (5% percentile to 95% percentile) of the endpoint environmental impact values, resulting from 1000 MC simulations have been illustrated as the upper and lower error bars in the graph of environmental impacts.

2.5 Developing parametric life cycle inventories (LCI) for environmental sensitivity analysis

Sensitivity analysis determines how big the outcomes of a model are sensitive to the variations of the input parameters and unveils the relative importance of the input parameters. As a prerequisite for sensitivity analysis, a parametric model is needed to be developed through incorporating the input parameters in the model (Table S2), mathematically linked together and to the outputs (Soleimani and Gilbert, 2020). Based on Eq.1, Eq.3 and Eq.4, experimental data from the lab and the literature, the parametric LCIs were developed in openLCA for 'one-at-a-time (OAT)' sensitivity analysis of the environmental impacts. In the OAT sensitivity analysis, the relative change in the environmental impacts due to $\pm 10\%$ relative change in the target parameter while all other parameters held constant, were calculated. To develop the life cycle inventory of the phytoremediation processes and

valorization scenario, all the materials consumed, and the process carried out including the preparation of the site, plant cultivation and harvesting until the end of the valorization of the biomass, were included.

2.6 Cost and revenue evaluation

A complementary economic evaluation parallel to the environmental assessment of phytoremediation alone and combined with the biomass valorization chain, provides insight into the viability of scenarios for decision-makers. Accordingly, the Cost Revenue Analysis was conducted in Excel based on the aforementioned equations of 1 to 8, information in the literature as well as the market prices in France. The average cost of €1200/ha (Jámbor and Török, 2019), and €700/ha (Pilu et al., 2013), for the cultivation of *A. donax* on an annually basis are reported in the literature for a 15 to 20-year life span. The cost of electricity generation with the gas turbine in combined heat and power (CHP) systems is estimated to be 0.1436 \$/kWh, and the cost of heat generation is estimated to be 0.0738 \$/kWh (Kang and Cao, 2014; Abanades et al., 2022). A range of 0.05\$/kWh to 0.18\$/kWh was reported for electricity generation in gas turbines based on the volume fraction of methane in biogas, the size of powerplant and the price of biogas (Barzegaravval et al., 2018).

The cost of 1m³ sediment phytoremediation per year was calculated based on assuming 5000 euros cost for one hectare of *A. donax* per year and the cost of 1 kWh renewable electricity from biogas in a CHP was assumed to be 0.13 €/kWh. The revenue per kWh electricity of 0.18 €/kWh, adopted from an average price of French electricity suppliers in 2023 considered as a negative cost.

3 Results

In this study, a laboratory-based experiment was integrated into economic and environmental life cycle assessment to offer a holistic approach to assess the impacts of phytoremediation combined with biomass valorization. A series of experiments were set up to study the feasibility of chloride phytoremediation by *A. donax* as well as its chloride bioaccumulation performance. The results of economic and environmental assessments and the laboratory trials are reported.

3.1 Experimental assessment of chloride phytoremediation performance of *A. donax*

The feasibility of phytoremediation of contaminated chloride-rich sediment by *A. donax* and its bioaccumulation performance was assessed through cultivation, growing and treatment trials. The deposition rates of chloride in aerial parts of *A. donax* in sediment treated separately with humic acid (8921mg/kg) and mycorrhiza (8221 mg/kg) were obtained lower than the deposition rate in non-treated sediment (9940 mg/kg). However, the sediment treated with combined humic acid and mycorrhiza resulted in more than a 50% increase in chloride deposition rate (15192 mg/kg) in aerial parts of *A. donax*. The reasons of the inhibitory and excitatory roles of separate and combined humic acid and mycorrhiza are not in the scope of this study. The mean deposition rate of chloride in aerial parts of *A. donax* cultivated in non-treated sediment, 9940 mg/kg dry biomass, was retained for the estimation of the required time frame of phytoremediation (Fig. 3).

3.2 Biomass yield and duration of phytoremediation versus sediment depth

Since the significant part of the root system of perennial grasses is mostly developed in the 0-15 cm upper surface of the soil (Gist and Smith, 1948; Rabêlo et al., 2021), the sediment depth (H) is very important in the performance of grass-based phytoremediation. On the other

side, according to equations 1 and 2, the sediment depth (H) is a key parameter to determine the plant density in a volume of sediment (plant/m^3), the estimation of the time frame of phytoremediation and the dry biomass yield per m^3 of sediment. The variation in the phytoremediation time interval and the dry biomass yield of *A. donax* versus the variations in the sediment depth (H), based on equations 1 and 2, are visualized in Fig. 3.

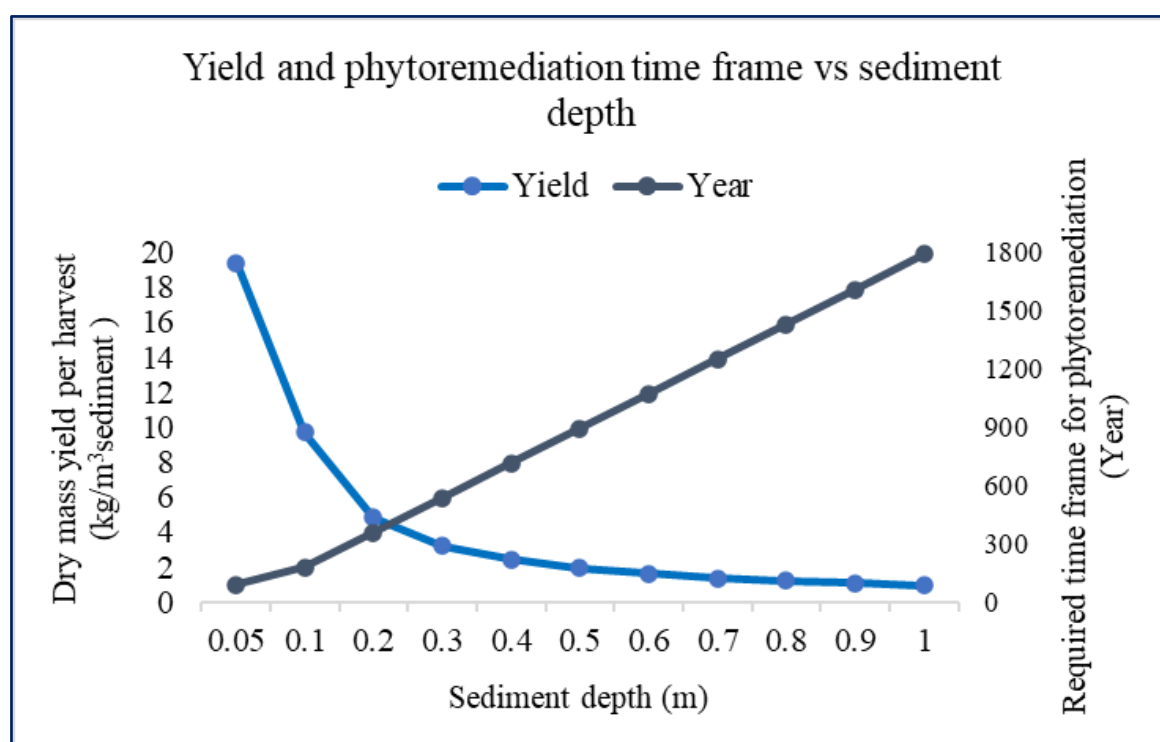


Fig. 3. Visualized linear relationship between the sediment depth and time frame of chloride phytoremediation (year) and inverse relationship between the sediment depth and biomass yield ($\text{kg}/\text{m}^3 \text{ sediment}$)

Assuming 20 cm sediment depth, the graph in Fig. 3 reveals that lowering the sediment chloride level from the current concentration of 16000 mg/kg up to the target regulatory concentration of 800 mg/kg, would not be achievable in less than 357 years.

3.3 Comparative environmental impacts of phytoremediation scenarios

The impacts of phytoremediation of 1m³ sediment, alone and integrated into green electricity generation in a CHP power plant, are compared in 18 impact categories of ReCiPe midpoint in Fig. 4, on a relative percentage basis.

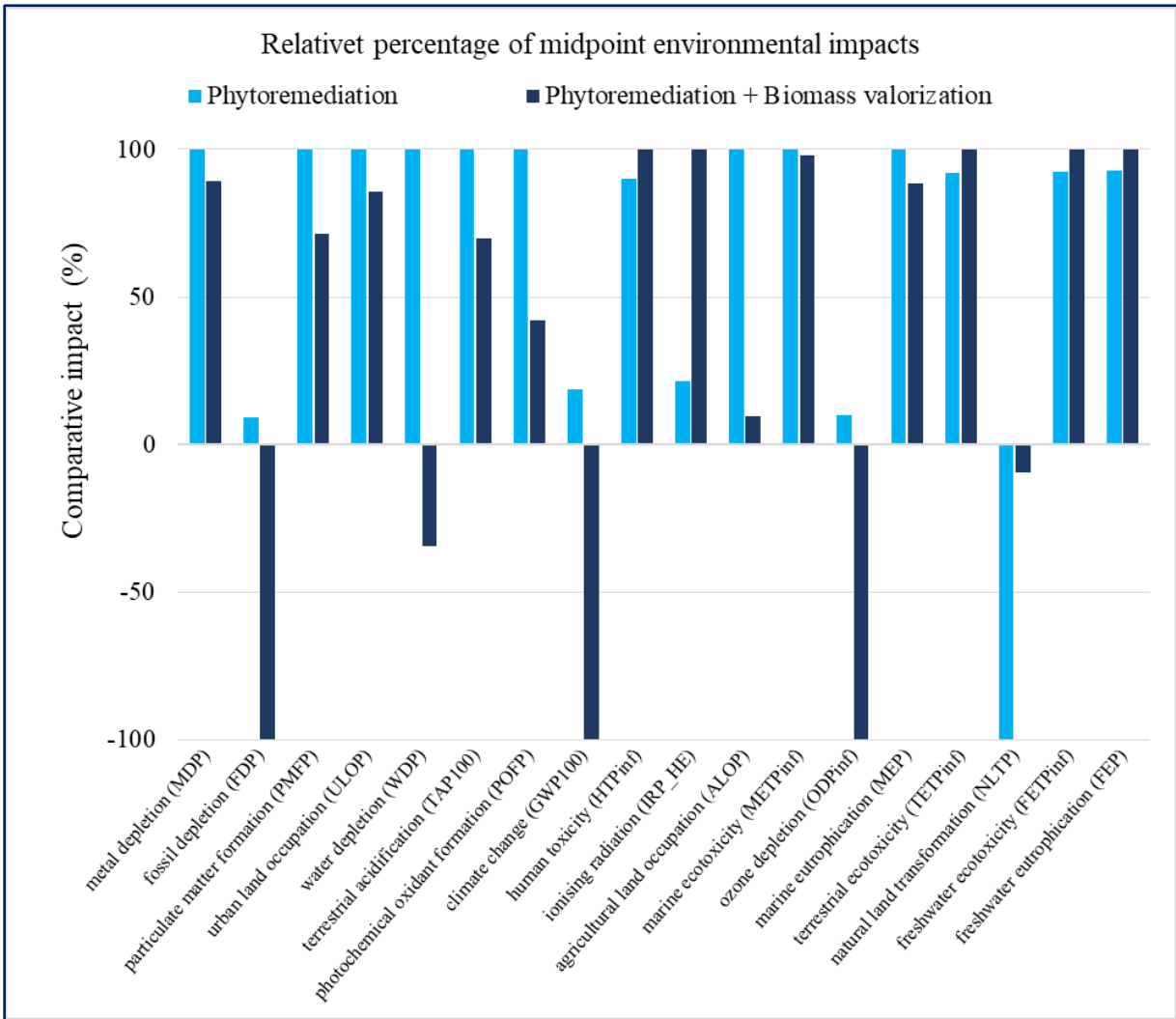


Fig. 4. Comparative environmental impacts of phytoremediation of 1m³ sediment, alone and integrated with biomass valorization to renewable electricity, in 18 impact categories of ReCiPe midpoint (H), based on relative percentage.

It is apparent that the phytoremediation scenarios could be distinguished by negative quantities for midpoint impacts of fossil depletion, water depletion, climate change and ozone

depletion. To have an idea about the final impacts on ecosystem quality, resources and human health, the endpoint impacts of phytoremediation scenarios were obtained in a dimensionless score of point (Pt) from the ReCiPe endpoint method. The endpoint impacts of the scenarios, normalized to the corresponding of phytoremediation scenario without biomass valorization, are visualized in Fig. 5 to provide comparative insight into the difference between the scenarios at the endpoint level.

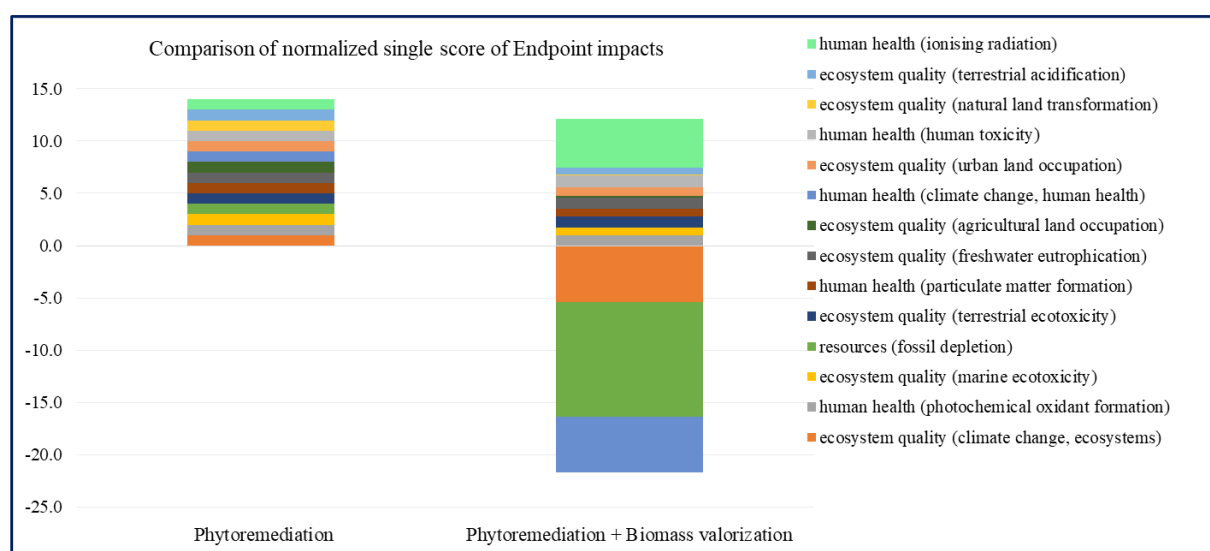


Fig. 5. Comparative normalized endpoint environmental impacts of sediment phytoremediation scenarios obtained from ReCiPe endpoint method.

At the endpoint level, the scenarios could be distinguished by negative impacts in categories of resources (fossil depletion), ecosystem quality (climate change) and human health (climate change).

3.4 Sensitivity analysis of environmental impacts of 1m³ sediment phytoremediation integrated with biomass valorization

The parametric LCIs enabled to perform the sensitivity analysis on intended parameters, were incorporated in the models. The sensitivity of local and global midpoint environmental

impacts of climate change, fossil depletion, freshwater eutrophication, human toxicity, and terrestrial acidification conducted in openLCA, the results are illustrated in separate graphs in Fig. 6. Each graph shows the variation percentage in the environmental impact due to a $\pm 10\%$ change of the targeted input parameters of sediment depth, the energy conversion efficiency of the bioenergy power plant, distance to the bioenergy power plant, biogas yield and plant survival rate.

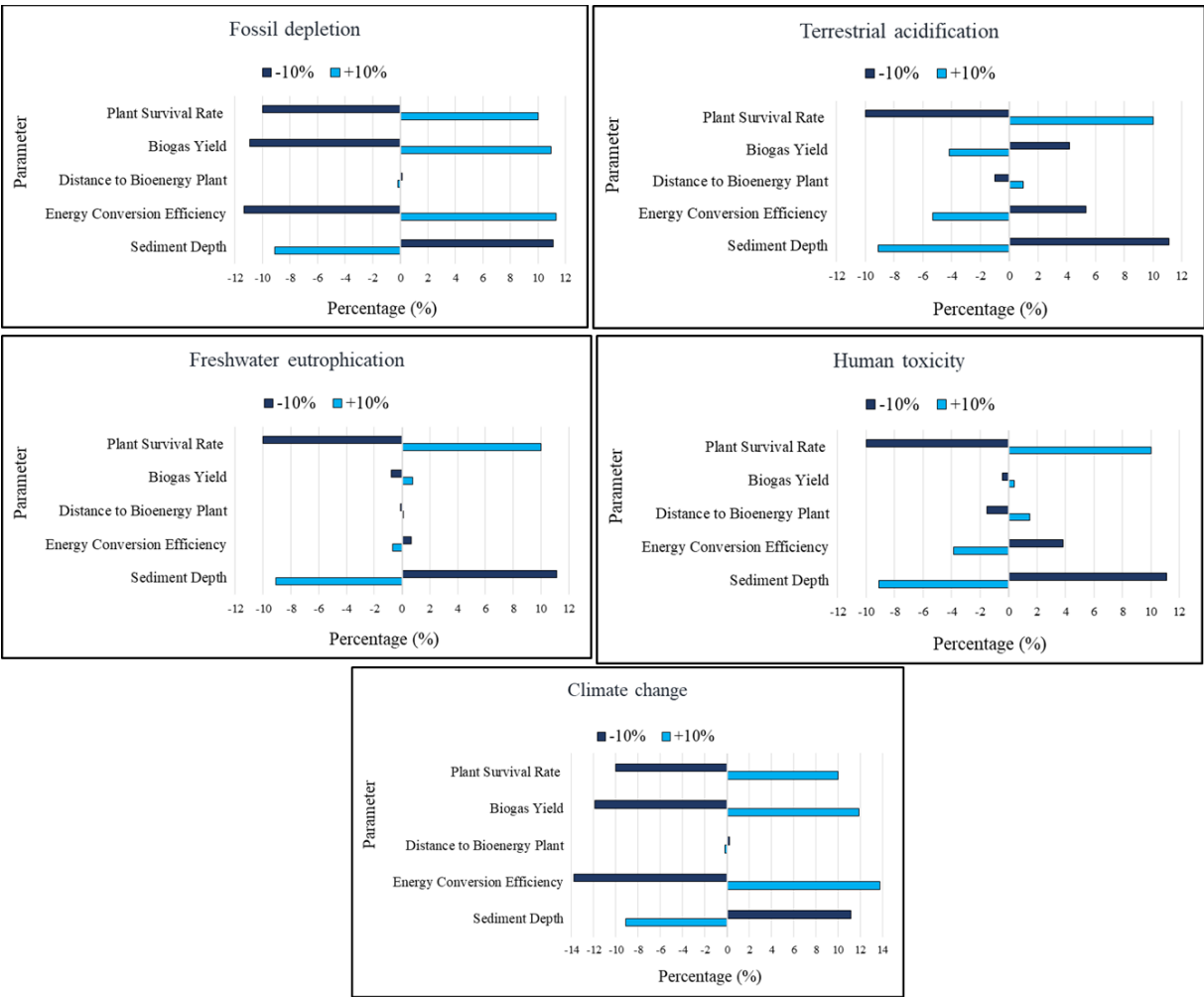


Fig. 6. Variation percentages in the environmental impacts of phytoremediation of 1 m^3 sediment integrated with biomass valorization in midpoint local and global impact categories of climate change, fossil depletion, freshwater eutrophication, human toxicity, and terrestrial acidification, due to $\pm 10\%$ one-at-time change of the target input parameters.

The graphs illustrate that climate change and fossil depletion have the maximum sensitivity to the efficiency of the power plant and biogas yield as was expected. The impact categories have the minimum sensitivity to the distance from the site to the bioenergy power plant, except for human toxicity. Relatively high sensitivity to sediment depth and plant survival rate is common between all the impact categories.

3.5 Uncertainty analysis of environmental impacts of sediment phytoremediation scenarios

The uncertainty analysis takes into account the uncertainty in all the parameters incorporated in the model to determine the lower and upper bands in each impact category with 90% confidence. The Monte Carlo simulation was carried out in openLCA for both phytoremediation scenarios to unveil the uncertainties in the endpoint impact categories of Human health, Resources and Ecosystem quality. The uncertainties in the endpoint impacts obtained from 1000 runs of Monte Carlo simulations are visualized as the error bars in Fig. 7. The upper band represents the 5% percentile, the lower band represents the 95% percentile and the range between them represents 90% confidence interval in each impact category.

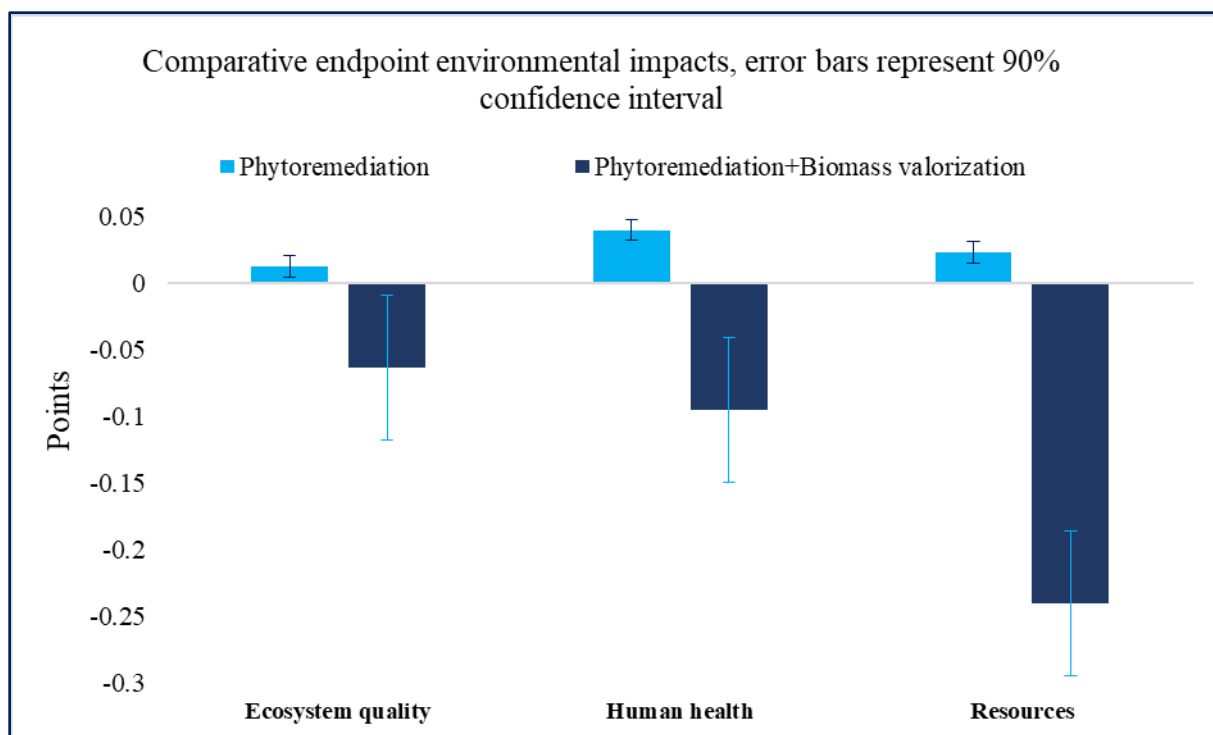


Fig. 7. Comparative endpoint environmental impacts of phytoremediation of 1m³ sediment, alone and combined with biomass valorization, along with the upper and lower error bars representing 5% percentile and 95% percentile and the interval of 90% confidence in each impact category.

3.6 Comparative economic and environmental impacts assessment of sediment phytoremediation scenarios

To provide a comparative overall view for the decision-makers on the economic and environmental costs of the scenarios, the 18 midpoint environmental impacts, normalized to the corresponding of phytoremediation were integrated into a single score. The costs of both scenarios were normalized to the corresponding of phytoremediation as well. To compare the sustainability of scenarios, the mean values of singles score environmental impacts, and the normalized costs are superimposed in a single graph in Fig. 8.

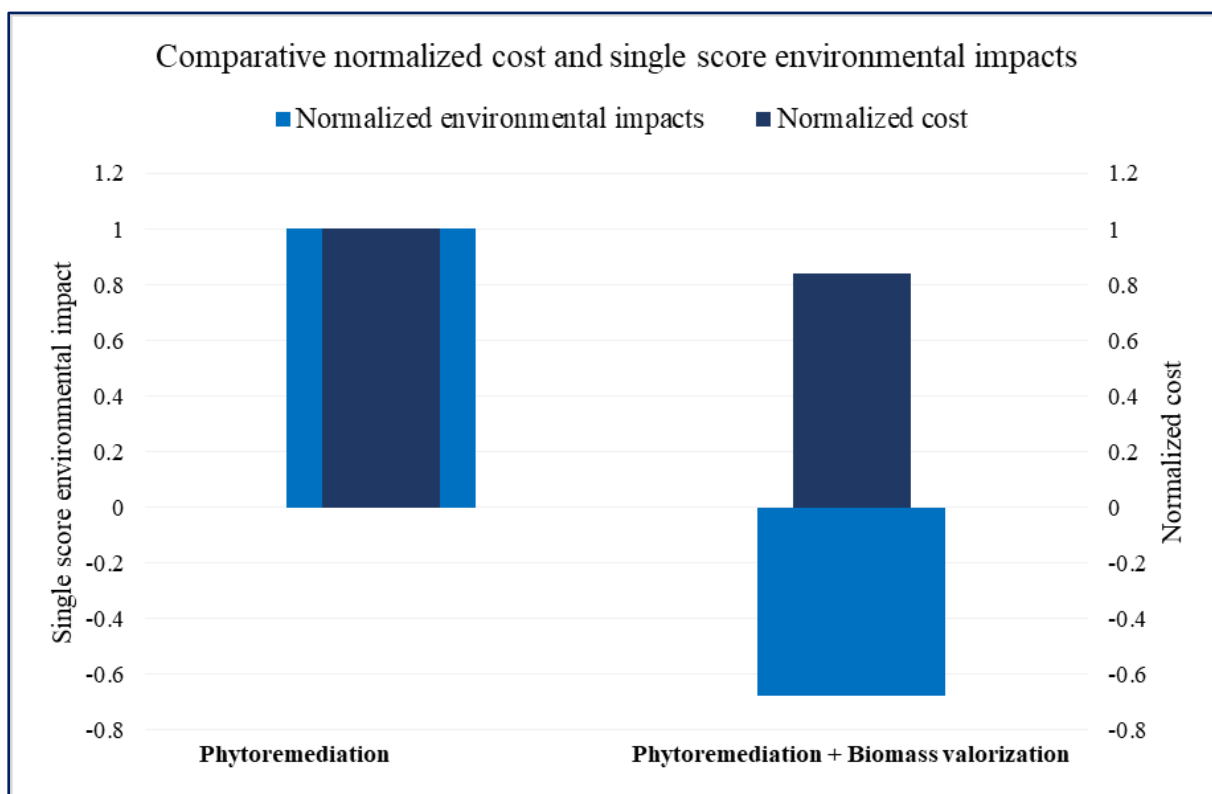


Fig. 8. Singles score environmental impacts and cost of scenarios, normalized to corresponding of phytoremediation.

The graph in Fig. 8 illustrates that the scenario of phytoremediation integrated with biomass valorization to bioenergy would have a relatively negative overall environmental impact compared to the phytoremediation scenario. From an economic perspective, inferred from the graph, biomass valorization could partially compensate for the costs of sediment phytoremediation. The rate of cost compensation depends on biomass yield per hectare, the market price of bioenergy, and the efficiency of the biomass valorization chain. In order to compare the costs and environmental impacts of other sediment management approaches with those of phytoremediation, sediment landfill was adopted for elaboration as a conventional alternative. The environmental impacts of sediment landfill, based on the sediment composition analysis (Table S1), obtained in 18 impact categories from the same method of ReCiPe midpoint (H), were normalized separately to those of phytoremediation scenarios.

The summation of unitless values were divided to corresponding of phytoremediation scenarios to obtain a single score. The cost of a 1m³ sediment landfill in the French market, 100 euros was adopted as the cost of a landfill and was normalized separately to corresponding of the phytoremediation scenarios. The means of single score impacts as well as the normalized cost of sediment landfill in France, are superimposed in a single graph to offer insight into the difference in the order of magnitude between the cost and impacts of 1m³ sediment landfill and those of annual sediment phytoremediation scenarios.

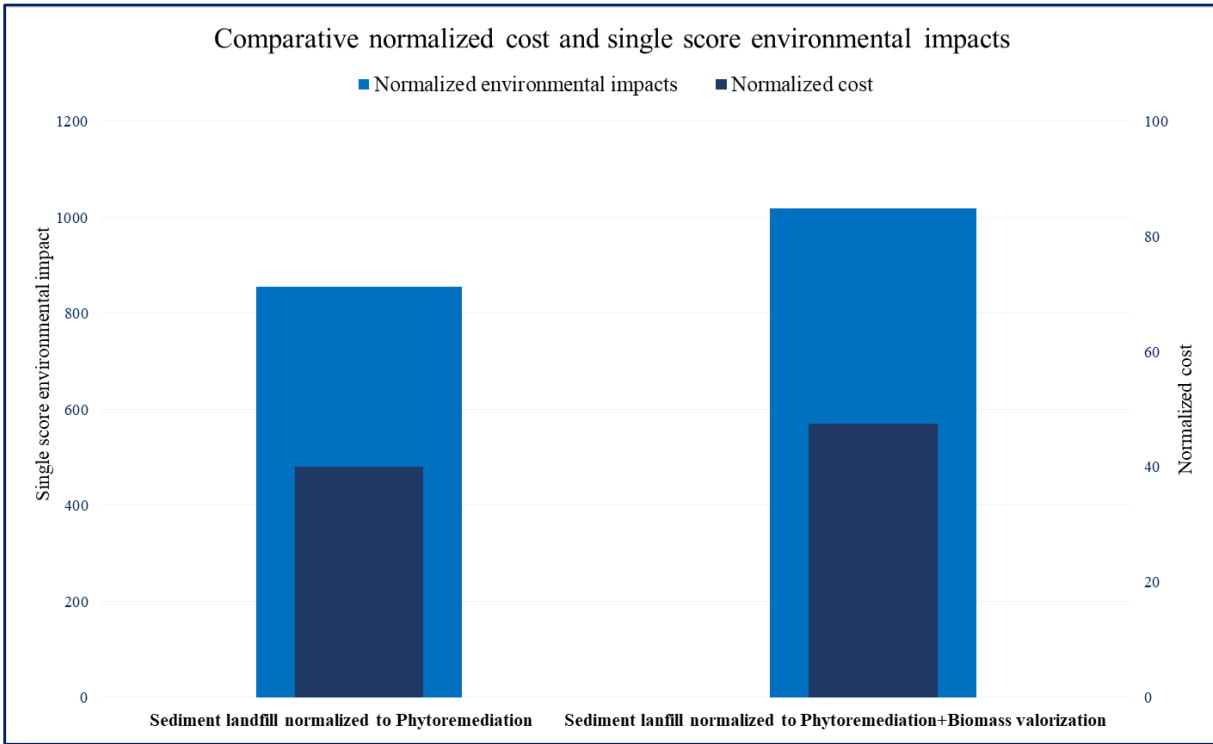


Fig. 9. Environmental impacts and cost of 1m³ sediment landfill, normalized to corresponding phytoremediation scenarios, integrated to singles scores are compared along with the normalized costs.

The graph shows that the environmental impacts of sediment landfill could be hundreds of times larger than the annual impacts of phytoremediation scenarios, mainly due to the leaching and emission of multiple trace elements and inorganic pollutions into air, water, and

soil. In terms of economy, the cost of a sediment landfill could be tens of times bigger than the annual cost of phytoremediation scenarios.

4 Discussion

The concentration of leachable chloride as a common element in all marine dredged sediments generally exceeds the regulatory threshold value. From the laboratory trials of growing *A. donax* in non-treated sediment and treated with Humic acid and Mycorrhiza, its tolerance to multi-element pollutants and chloride-rich sediment was confirmed with zero mortality rate of cultivated plants. In terms of the chloride bioaccumulation performance, the experiments unveiled that the accumulation capability of *A. donax* is considerable enough to be categorized among hyperaccumulator plants, which is generally referred to the plants with accumulation capability of more than 1000 mg/kg dry mass (Roy et al., 2005). Based on these results, conducting phytoremediation for chloride-rich sediment via *A. donax* is feasible. However, the estimated time frame of phytoremediation showed that due to the high concentration of chloride in sediment compared to the regulatory value to be considered as an inert material (800 mg/kg), phytoremediation of the sediment dredged from the port of Camargue is not practical in a reasonable duration of time.

According to the inverse relationship between phytoremediation time frame and the chloride deposition rate of *A. donax* (d_r), chemically assisted increase in d_r up to 15192 mg/kg dry mass by combined application of Humic Acid and Mycorrhiza would decrease the time frame to 234 year which is not applicable as well. To have the chloride inert sediment in a reasonable time frame of 20 years, based on the equation 2, the chloride concentration should be decreased from the current quantity of 16000 mg/kg up to 1650 mg/kg, which might be

achievable through complete sediment dewatering in geotube or sediment washing during a dredging operation on the sea, or any other appropriate pretreatment. Although phytoremediation, unlike other conventional physicochemical remediation techniques, is categorized among the green technologies (Nedjimi, 2021), like any other agricultural activity has environmental impacts as is apparent in the positive values of 17 midpoint impact categories. Considerable negative midpoint and endpoint environmental impacts emerged when the valorization of biomass to green electricity was integrated into phytoremediation.

Because of the importance of considering sediment depth in the effectiveness of plant rooting system as well as estimating the time frame, the surface area may not be an appropriate functional unit in the phytoremediation LCA studies. In spite of the recommendations to consider one hectare as a suitable functional unit for phytoremediation LCAs (Morais and Delerue-Matos, 2010; Vigil et al., 2015) the more appropriate functional unit seems to be 1m^3 of sediment: 1) to incorporate the sediment depth into the model, 2) to consider the effectiveness of plant rooting system, 3) to estimate the phytoremediation time frame and 4) to retain economic and environmental comparability to other valorization or landfill scenarios usually are expressed on volume or mass basis, and 5) to estimate harvested biomass per m^3 of sediment based on plant cultivation density.

A sensitivity analysis was carried out to identify how variations in the input parameters contribute to the targeted local and global impacts to determine the key parameters in each impact category. The sensitivity analysis unveiled that the parameters involve in biomass yields per 1m^3 sediment such as the sediment depth and the survival rate of plants as well as the parameters involve in ultimate energy yields such as the biogas yield and the efficiency of the bioenergy power plant are the key parameters affecting the environmental impacts. While

sensitivity analysis determines the relative importance of the input parameters in model outputs, uncertainty analysis unveils the entire possible outcomes together with their level of certainty represented by the confidence interval. The 90 percent confidence interval means that considering all uncertainties in the model, the impacts would be in the confidence interval with the probability of occurrence of 90 percent. According to the results of uncertainty analysis, the uncertainty in the impacts of phytoremediation integrated with biomass valorization is higher than the corresponding of phytoremediation, but none of the uncertainties is large enough to change the game between the scenarios.

The comparative economic and environmental impacts of sediment phytoremediation scenarios, demonstrated that biomass valorization could offset the environmental impacts and partially compensate the costs of phytoremediation, offering a more viable sediment remediation scenario. The mitigation of the environmental impacts could mainly be attributed to carbon sequestration and the avoided fossil fuels for electricity generation. Since the values for biomass yield, bioenergy yield, power plant efficiency, etc., considered in life cycle assessments are conservatively on an average basis, higher benefits in terms of economy and environment could be expected from higher biomass yield in cropping conditions and improvements in the biomass valorization chain. In addition to the improvements and optimizations, the regular hike in the price of fossil fuels as the main competitor for renewable bioenergy would offer more desirability for biomass valorization in terms of economy. The solid digestate remainder of anaerobic digestion which is nutrient-rich and comparable to manure could be applied to replace chemical fertilizers (Abanades et al., 2022). As far as the environmental and economic profits of the solid digestate are not considered in this study, additional benefits would be expected from biomass valorization.

The costs and environmental impacts of sediment landfill, as an inevitable alternative scenario for phytoremediation, compared to corresponding of phytoremediation scenarios, demonstrates that phytoremediation of 1m³ sediment would be relatively much higher viable than the landfill, even without biomass valorization. In comparisons between the landfill and phytoremediation of sediment, the time frame is compromised because of the long-term nature of phytoremediation. In case the phytoremediation of non-inert chloride-polluted sediment is not achievable in a reasonable and realistic time frame, phytoremediation could be considered as a pretreatment for other sediment valorization scenarios such as substitution in cementitious materials (Soleimani et al., 2023) or marine grade concrete (Hayek et al., 2023) after partial remediation of detrimental effects of pollutants.

5 Conclusion

A mathematical model was developed to estimate the maximum time frame for phytoremediation of multiple-pollutant sediment as well as the yield of biomass to bioenergy valorization chain including the estimation of biomass yield per m³ of sediment, the bioenergy yield from valorization schemes and final green electricity per m³ of sediment, as a prerequisite for economic and environmental assessments. The mathematical model was applied to develop parametric life cycle inventories for two scenarios of phytoremediation separately or integrated with biomass valorization. A laboratory-based experiment was integrated into economic and environmental life cycle assessment to offer a holistic assessment approach.

From the comprehensive economic, environmental, and experimental assessments, the following implications could be inferred:

- Integrating biomass to bioenergy valorization chain into sediment phytoremediation would offer superior viability in terms of economy and environment.
- The sediment depth is very important for the effectiveness of the plant root system in phytoremediation, as well as the estimation of the time frame and the biomass yield per m³ sediment.
- Volume of sediment (1m³) is a more appropriate functional unit for LCAs of phytoremediation than the surface area (1ha) in order to incorporate the sediment depth into the model to consider the plant rooting system and retain economic and environmental comparability to other valorization scenarios.
- The phytoremediation of non-inert chloride-polluted sediment at the current state of the art may not be a short-term strategy achievable in a reasonable and realistic time frame.
- The model could be evolved more, through the incorporation of other targeted influential parameters and their uncertainties to provide comprehensive insight into the affecting parameters on the economic and environmental impacts.
- Phytomanagement of dredged sediment could be carbon negative scenario thanks to carbon sequestration and avoiding fossil fuels.
- As a recommendation for future work, further economic and environmental assessments for other biomass valorization schemes integrated into the

phytoremediation, could enlighten more the perspective of sediment
phytoremediation.

- As potential areas for future investigations, the model developed in this study is a general model applicable for different types of plants, contaminated sediment and soils, and biomass to bioenergy valorization schemes.

CRedit authorship contribution statement

Not applicable.

Data availability

Data will be made available on request.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online.

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