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Biochar produced from wood waste for soil remediation in Sweden: Carbon sequestration and other environmental impacts

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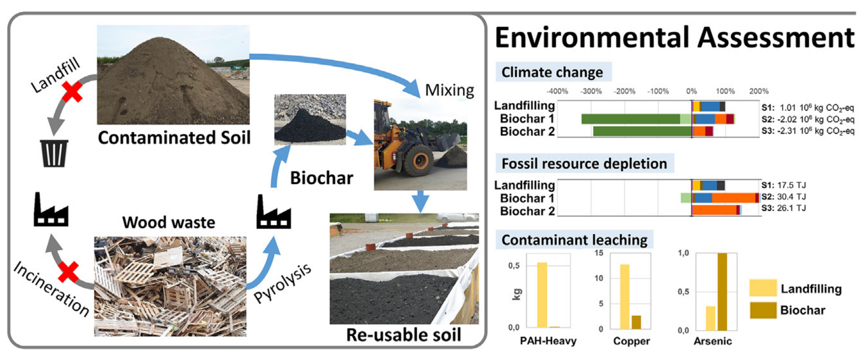
HIGHLIGHTS

- Using biochar for soil remediation was compared to landfilling of contaminated soil.
- Biochar treatment brought large reductions of CO_2-emissions compared to landfilling.
- 100-year leaching of PAH and Cu was much lower in treated soil than in landfill.
- 100-year leaching of several other metals was too sensitive to model assumptions.
- Biochar for remediation is promising but needs case-specific risk assessment.

GRAPHICAL ABSTRACT



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ABSTRACT

The use of biochar to stabilize soil contaminants is emerging as a technique for remediation of contaminated soils. In this study, an environmental assessment of systems where biochar produced from wood waste with energy recovery is used for remediation of soils contaminated with polycyclic aromatic hydrocarbons (PAH) and metal(loid)s was performed. Two soil remediation options with biochar (on- and off-site) are considered and compared to landfilling. The assessment combined material and energy flow analysis (MEFA), life cycle assessment (LCA), and substance flow analysis (SFA). The MEFA indicated that on-site remediation can save fuel and backfill material compared to off-site remediation and landfilling. However, the net energy production by pyrolysis of wood waste for biochar production is 38% lower than incineration. The LCA showed that both on-site and off-site remediation with biochar performed better than landfilling in 10 of the 12 environmental impact categories, with on-site remediation performing best. Remediation with biochar provided substantial reductions in climate change impact in the studied context, owing to biochar carbon sequestration being up to 4.5 times larger than direct greenhouse gas emissions from the systems. The two biochar systems showed increased impacts only in ionizing radiation and fossils because of increased electricity consumption for biochar production. They also resulted in increased biomass demand to maintain energy production. The SFA indicated that leaching of PAH from the remediated soil was lower than from landfilled soil. For metal(loid)s, no straightforward conclusion could be made, as biochar had different effects on their leaching and for some elements the results were sensitive

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to water infiltration assumptions. Hence, the reuse of biocharremediated soils requires further evaluation, with site-specific information. Overall, in Sweden's current context, the biochar remediation technique is an environmentally promising alternative to landfilling worth investigating further.

1. Introduction

Contamination of soils from human activities is one of the most significant environmental problems in the contemporary world (Zama et al., 2018). Only in Europe, it has been estimated that there are about 2.8 million potentially contaminated sites in EU-28 (Pérez and Eugenio, 2018). An extensively used method for handling soil contaminated with e.g. heavy metals and persistent organic pollutants is to excavate and landfill the contaminated soil, and then backfill the excavated area with clean material. However, this technique is problematic due to the scarcity of landfill space, high energy requirements, and limited availability of natural resources for backfill.

A wide range of remediation techniques have been developed, such as bioremediation, chemical oxidation, steam injection, or soil washing (de Albergaria and Nouws, 2016). These techniques are usually optimised for one or a few specific contaminants, and can be implemented either on-site or off-site (Busset et al., 2012; Lemming et al., 2010a). The cost and environmental impacts of remediation techniques vary widely (Amponsah et al., 2018), but most commonly arise from energy-intensive processes, use of chemicals, and long transport distances (Suèr et al., 2004). Efforts are being made to develop new, more efficient, remediation techniques, with lower costs and lower environmental impacts.

One technique under development is the use of biochar to stabilize soil contaminants. Biochar is the solid carbonaceous product obtained from the thermochemical conversion of biomass in an oxygen-limited environment (e.g. pyrolysis) (Moreira et al., 2017). Biochar is now recognized as a CO_2 removal technology (IPCC, 2019), as producing biochar and mixing it with soil is an effective solution for sequestering atmospheric CO_2 (Lehmann and Joseph, 2009). Beyond carbon sequestration, multiple uses of biochar are being developed across various sectors, owing to its versatile properties (Nanda et al., 2016). The sorption properties of biochar, high surface area, and ion exchange capacities have spurred an interest in using biochar for remediation of contaminated soils with organic (e.g. polycyclic aromatic hydrocarbons; PAH) and/or inorganic compounds (e.g. metals and metalloids; metal(loid)s) (Yuan et al., 2019).

Several studies have reported the potential of biochar for remediating contaminated soils with organic and/or inorganic pollutants (Beesley et al., 2011; Paz-Ferreiro et al., 2014; Yuan et al., 2019; Zama et al., 2018). The efficacy of biochar for soil remediation depends on its properties, which in turn depend on the biomass feedstock and the production conditions (e.g. temperature, retention time) (Weber and Quicker, 2018). For hydrophobic organic compounds like PAH, sorption onto biochar improves with higher pyrolysis temperature, as the biochar surface is increased and becomes more aromatic and hydrophobic (Hassan et al., 2020; Zama et al., 2018). Sorption of inorganic compounds is more complex and dependent on soil and pore-water properties, and the surface chemistry of the biochar. The main sorption mechanism is due to cation exchange reactions with the biochar surfaces. Good results of heavy metal sorption have thus been reported for elements like Cd, Cu, Pb, Zn, that exist as positively charged ions in soil pore water (Beesley and Marmiroli, 2011; Cárdenas-Aguiar et al., 2017; Thomas et al., 2020). However, immobilization of negatively charged metal(loid)s, like As Sb and Mo, often has proven ineffective or even counter-productive with increasing pore water concentrations of these elements (Hilber et al., 2017; Zama et al., 2018). In case that PAH and metal(loid)s co-exist in multicontaminated soils, competition phenomena between different contaminants for sorption sites may occur, and the sorption efficacy of biochar varies depending on the biochar, soil, contaminant concentrations, and contaminants themselves (Cao et al., 2009; Janus et al., 2020; Kołtowski and Oleszczuk, 2016; Kong et al., 2011). Hence, the use of biochar for soil remediation requires case-specific assessments, as there may be trade-offs, especially in the case of soils with multiple contaminants (Beesley et al., 2011).

In Sweden, the dominant technique for remediation of contaminated sites is still excavation and landfilling of soil, commonly referred to as “dig and dump” (Anderson, 2017). This technique also includes the need of virgin soil materials for backfill at the excavation sites. Soil is a valuable and limited resource with a very low rate of regeneration that must be managed in a sustainable way (McBratney et al., 2014). Thus, it is important to find more sustainable remediation techniques. In this perspective, the Biochar-RE:Source project (Enell et al., 2020) was set up to develop and test a remediation technique based on biochar made from urban wood waste. The project studied biochar remediation of contaminated soil from the urban area of Helsingborg, Sweden, and included contaminant leaching experiments of treated and untreated soil.

Remediation of contaminated soil using biochar is a relatively new technique and, to our knowledge, its environmental performance has not been fully assessed. Several studies used Life Cycle Assessment (LCA) to analyze the environmental impacts of systems where biochar, produced from pyrolysis of biomass, is applied to soils (for a comprehensive review see Matuštík et al. (2020)). However, the produced biochar in these systems is used as soil amendment in agricultural soils. It is therefore necessary to assess potential environmental impacts and other environmental aspects of systems where the produced biochar is used as a remediation technique to stabilize soil contaminants.

LCA is the most widely applied tool to assess the environmental sustainability of different products or services. It is a systematic method for assessing the environmental impacts of a product throughout its whole life cycle, which through its holistic perspective, can prevent problem shifting between different life cycle stages, impacts, or regions (Ita-Nagy et al., 2020). Material Flow Analysis (MFA) is important for LCA as it forms the basis for the LCA modeling (Guo et al., 2021). MFA is the systematic assessment of the flows and stocks of materials within a system defined in space and time, aiming to connect the sources, the pathways, and the sinks of materials in the system (Brunner and Rechberger, 2016). MFA is often referred to as Material and Energy Flow Analysis (MEFA) if its scope is expanded to include also energy flows (Naohiro et al., 2015) or as Substance Flow Analysis (SFA) if specific substances are the focus (Brunner and Rechberger, 2016). Apart from supporting LCA modeling, the application of MEFA and SFA offers insight into the functioning of the assessed systems and allows a transparent and open way to inform researchers and external stakeholders (Allesch and Brunner, 2017). SFA can also complement LCA, as it can provide a detailed quantitative description of substance flows throughout a system, which LCA cannot provide due to aggregation of flows at the impact level (Azapagic et al., 2007).

Given this background, the aim of this study is to assess the environmental impacts, from a life cycle perspective, of using biochar produced from wood waste to remediate soil contaminated with PAH and metal(loid)s. Two different options of soil remediation with biochar are considered (on- and off-site) and compared to the conventional “dig and dump” technique. The specific objectives are to: (i) describe the material and energy inflows and outflows of different systems for management of contaminated soil and energy recovery from wood waste; (ii) assess the life-cycle environmental impacts of these systems; and (iii) map and quantify the flows and stocks of PAH and metal(loid)s present in the contaminated soil.

2. Methods

Four methodological steps were followed. First, three scenarios were developed to describe different systems for managing contaminated soil and wood waste at the studied site. Second, a MEFA was conducted in order to map and quantify the material and energy flows of the systems. Third, a LCA was conducted for assessing and comparing the environmental impacts of the systems. Finally, a SFA was carried out to analyze the flows and stocks of the contaminants.

2.1. Scenario definition

2.1.1. Study site

The study site is the waste management (WM) facility at Filborna in Helsingborg, southern Sweden. The facility is operated by the municipal company Nordvästra Skånes Renhållnings AB (NSR AB). Two of the waste streams managed at the site are urban garden waste and soil contaminated with metal(loid)s and PAH. The contaminated soil comes from excavation works in Helsingborg and is currently disposed in the landfill of the facility. The leachate from the landfill is collected and treated on-site and then discharged in the Öresund strait. The garden waste is sorted through shredding and sieving in two fractions. The woody fraction (henceforth called wood waste) is transported 125 km north to Falkenberg, where it is combusted for district heating. The green fraction (green waste), consisting mostly of leaves and soil, is composted in windrows on-site. NSR has been exploring alternative treatment options for the wood waste and the contaminated soil. They have recently decided to invest in a new pyrolysis plant (3700 kW biomass input), developed by (Biogreen, 2020), for converting the wood waste into biochar and heat for district heating. For the biochar, different uses are considered. The main use studied here is its application to the contaminated soil and the subsequent reuse of the soil mix for different purposes outside the facility.

2.1.2. Scenarios

The three scenarios are described below, while their visual representations, along with the system boundaries of the LCA, are shown in Fig. 1.

2.1.2.1. Scenario 1 (S1): dig and dump

S1 depicts the current situation at the NSR site as described above. Excavated contaminated sites in Helsingborg are assumed to be backfilled with uncontaminated virgin material (crushed gravel).

2.1.2.2. Scenario 2 (S2): off-site remediation with biochar

In S2, the sorted wood waste is dried and then run through a chipper that turns it into woodchips (15–30 mm). The woodchips are then converted through slow pyrolysis into syngas and biochar. The syngas is combusted and part of the produced heat is used for drying the wood waste, while the rest is delivered to the local district heating network. Contaminated soil from Helsingborg is transported to the NSR site where it is mixed with the biochar, and uncontaminated soil is transported to the excavation sites for backfilling. It is assumed that 94% of contaminated soil is mixed with 6% biochar (*w*/w). This assumption was based on data from field experiments during the Biochar-RE: Source project, where tests with 3% and 6% biochar indicated that the latter can provide greater reductions in the leaching of PAH and Cu, Ni, Hg and Zn from the soil (Enell et al., 2020). Higher proportions of biochar (e.g. 10%) were not tested, as the addition of more biochar in soil could cause adverse effects on terrestrial ecosystem (Khan et al., 2015). After mixing, the remediated soil is transported to the surrounding area for reuse.

2.1.2.3. Scenario 3 (S3): on-site remediation with biochar

S3 is similar to S2. The main difference is that the produced biochar from the pyrolysis plant is now transported to the excavation sites where it is mixed with the contaminated soil (94% soil, 6% biochar; w/w). The remediated soil is then reused on-site as backfill material.

NSR is considering using biofuels for all machinery in the WM facility and transportation of materials. Thus, all machinery within the WM facility and trucks transporting wood waste to the incineration plant (S1) and biochar to the site of use (S3) are assumed to run on biodiesel (rapeseed methyl ester). All other machinery and trucks use diesel.

2.2. Material and energy flow analysis

The goal of the MEFA was to map and quantify material and energy flows and stocks in the systems described by the scenarios. The system boundaries of the analysis included the processes in the WM facility, the excavation or on-site remediation of contaminated soil and the transportation of materials between the processes. Collection and transportation of garden waste to the facility and composting of green waste were excluded as they did not differ between scenarios. Moreover, the leaching of contaminants from the landfilled contaminated soil or from the reused remediated soil was excluded from the system boundaries. Instead, the leaching of contaminants was assessed separately through an SFA (see Section 2.4). The time boundary of the analysis was annual.

The material and energy flows were determined based mainly on data collected through personal communication with the NSR staff. Data gaps were resolved using information from the literature and applying the mass and energy balance principles. In particular, whenever data on fuel and electricity consumption of machinery was not available, data from the Ecoinvent database (Wernet et al., 2016) was extracted. Likewise, data from Ecoinvent, along with information about transportation distances from the NSR staff, were used to estimate fuel consumption for materials transportation. The mass and energy balances of the systems in S1, S2 and S3 were visualized as Sankey diagrams using the online software Sankey Flow Show (2020).

2.3. Life cycle assessment

A comparative process-based LCA was performed using the LCA framework Brightway2 (Mutel, 2017) and its graphical interface Activity Browser (Steubing et al., 2020).

2.3.1. Goal and scope definition

2.3.1.1. Goal. The goal was to assess the environmental impacts of three systems for remediation of contaminated soil and wood waste treatment with energy recovery, using the defined scenarios (S1, S2 and S3).

2.3.1.2. System boundaries. The system boundaries of the LCA include the same processes as the system boundaries of the MEFA (see Section 2.2). Moreover, they include the impacts from capital goods, i.e. buildings, machinery and means of transportation, upstream impacts from the production and transportation of material used for backfilling, and downstream impacts from the final disposal of bottom-ash. The background modeling choices reflected the local conditions in Helsingborg in various ways, e.g. electricity mix, district heating production, transport processes. For the time boundary, a 100-year time horizon was selected for the impact assessment.

2.3.1.3. Functional unit. The systems described in S1, S2 and S3 are multifunctional, as they provide three different functions: treatment of wood waste, production of heat for district heating, and management of contaminated soils. The functional unit was set to 1 year of operation of the pyrolysis plant (800 kg h^{-1} dry wood, 1250 t yr^{-1} biochar). This functional unit equates to the treatment of 5650 t of wood waste for district

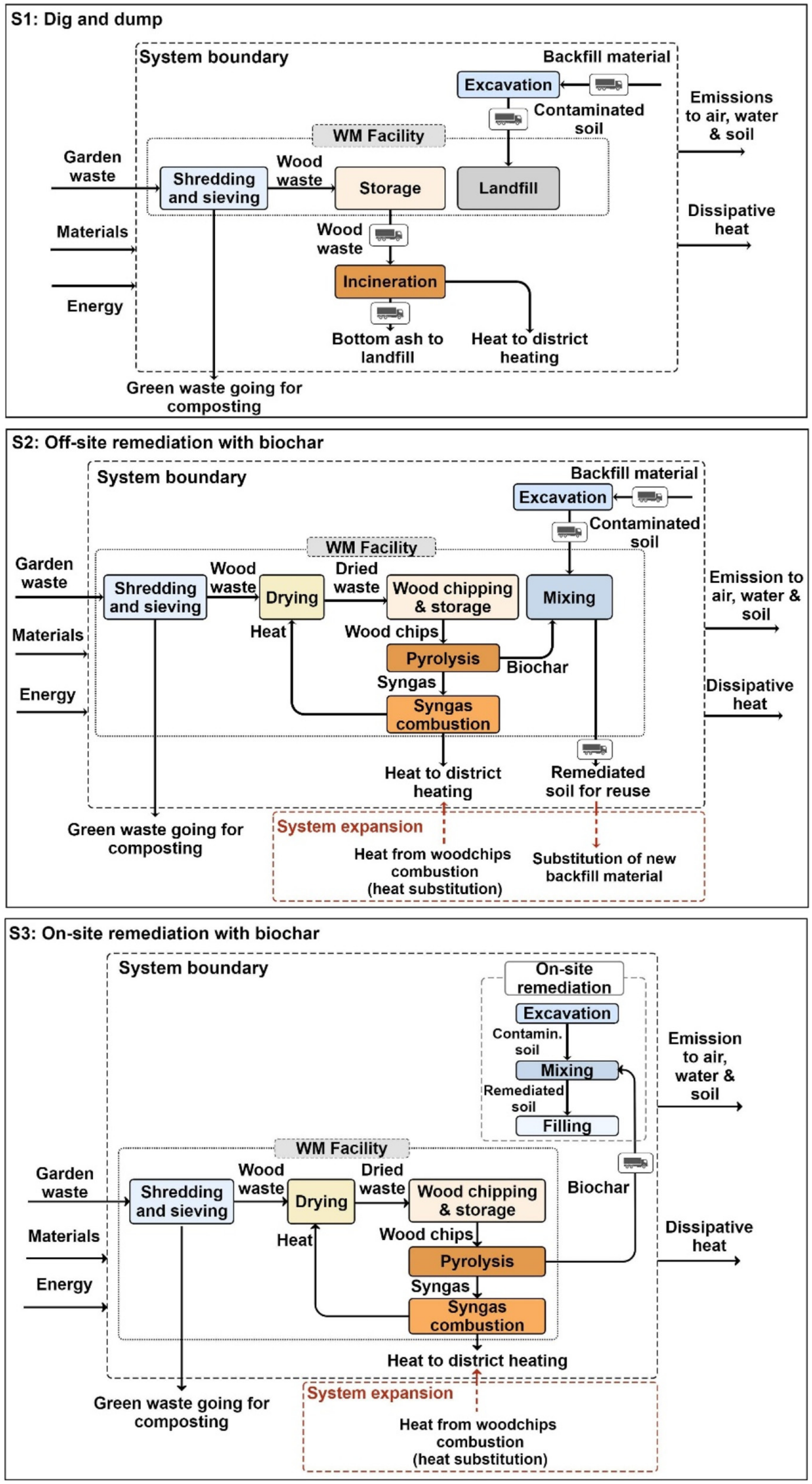

Fig. 1. System boundaries for LCA of S1, S2 and S3.

heating production and the management of 12,240 m^3 of contaminated soil. The choice of such a functional unit is common for multifunctional systems, like bio-refineries, as it allows to study the benefits of process integration (Ahlgren et al., 2015).

2.3.1.4. System expansion. The system expansion approach was chosen to keep the functional unit constant and ensure that different scenarios deliver the same services (ISO, 2006a, 2006b; Lausselet et al., 2017). S2 and S3 were expanded to include the production of heat from combustion of woodchips to compensate for the lower heat production from the pyrolysis plant (heat substitution). In addition, the remediated soil in S2 was assumed to be reused and substitute for inert uncontaminated materials used for backfilling. Thus, S2 included avoided burdens from the extraction and transportation of new materials for backfilling.

2.3.2. Life cycle inventory (LCI)

The LCIs of the three scenarios were compiled in the LCA software Brightway2 using the Ecoinvent version 3.6 (cut-off) database (Wernet et al., 2016). Several activities from the database were modified in order to better reflect local conditions. Moreover, new activities were modelled to simulate activities that were not available in the database. The modeling of the main activities is summarized in Table 1, while more details can be found in S.1 in Supplementary Material (S.M). The main assumptions made for the modeling are discussed below.

One of the effects of biomass pyrolysis is carbon sequestration in the biochar. The amount of sequestered carbon was estimated based on the approach followed by Azzi et al. (2019) assuming that the carbon stability of biochar in soil for the first 100 years is 80%. This in line with previous LCAs of woody biochars (Matuštík et al., 2020; Tisserant and Cherubini, 2019), meta-analysis of biochar incubation data (Wang et al., 2016) and the IPCC preliminary inventory methodology (IPCC, 2019).

In Sweden, there is a large potential for harvesting forest residues, i.e. tops and branches from forestry activities, and using them for bioenergy production (de Jong et al., 2017). Hence, it was assumed that heat from burning woodchips from forest residues compensates for the lower heat production in S2 and S3. It was also assumed that the harvesting of the residues does not cause land use change. However, if these residues were not harvested for bioenergy production, they would decompose over time in the forest contributing to forest carbon stocks. The climate change impact from loss in forest carbon stocks due to harvesting was estimated to be 89.1 kg CO_2-eq/t of woodchips (Azzi et al., 2019; Hammar et al., 2019).

Another assumption was that machinery in the WM facility and trucks transporting wood waste to incineration (S1) and biochar to the site of use (S3) run on biodiesel produced from rapeseed (rapeseed methyl ester), as NSR is considering to run all machinery and trucks on biodiesel. However, it is unknown what type of biodiesel will be used. It was assumed that the biodiesel is produced from rapeseed because of better data availability for this type of biofuel.

2.3.3. Life cycle impact assessment (LCIA)

The ILCD 2.0 impact assessment methodology (European Commission, Joint Research Centre, Institute for Environment And Sustainability, 2012) was applied. The following 12 midpoint impact categories were used: 1) Climate change, 2) Freshwater and terrestrial acidification, 3) Freshwater eutrophication, 4) Marine eutrophication, 5) Terrestrial eutrophication, 6) Ionizing radiation, 7) Ozone layer depletion, 8) Photochemical ozone creation, 9) Respiratory effects, inorganics, 10) Fossils, 11) Land use, and 12) Minerals and metals. Toxicity-related impact categories were excluded from the LCA, as the fate of metal(loid)s and PAH was studied separately in the SFA (see Section 2.4).

2.3.4. LCA sensitivity analysis

A sensitivity analysis was conducted to assess the influence of the assumption that machinery and trucks run on biodiesel. For this purpose, the scenarios were remodeled assuming that all machinery and transportation trucks run on diesel. Another analysis was performed to determine to what extent the electricity mix affects the results. The electricity mix in SE4, which was used in the scenarios, is mainly from renewables (hydro and wind) and nuclear. For the sensitivity analysis, the scenarios were remodeled assuming that the used electricity was from a fossil-based generation technology (natural gas – modelled using the Ecoinvent activity "Heat and power co-generation, natural gas, conventional power plant, 100MW electrical").

2.4. Substance flow analysis with a life cycle perspective

The goal of the SFA was to analyze the flows and stocks of PAH and metal(loid)s present in contaminated soil handled in the systems described in S1, S2 and S3. The system boundaries included the same processes as the system boundaries in LCA and, in addition, they included leaching of contaminants from landfilled contaminated soil in S1 and from reused remediated soil in S2 and S3. 12 metal(loid)s and 16 PAH were included in the analysis. The metal(loid)s are: As, Ba, Cd, Cr, Co, Cu, Pb, Hg, Mo, Ni, V and Zn. The PAH are: Acenaphthene, Acenaphthylene, Anthracene, Benz[*a*]anthracene, Benzo[*a*]pyrene, Benzo[*k*]fluoranthene, Benzo[*b*]fluoranthene, Benzo[*ghi*]perylene, Chrysene, Dibenz[*ah*]anthracene, Fluorene, Fluoranthene, Indeno[1,2,3-*cd*]pyrene, Naphthalene, Phenanthrene and Pyrene. The timeframe of the analysis was 100 years.

LCI emissions of substances from the processes in the modelled scenarios were extracted via the LCA software Brightway2. For the metal(loid)s, estimates of the amounts leaching from the landfilled contaminated soil and the reused remediated soil were derived using data from measurements with lysimeters that were installed in test beds with these soils. For the PAH, data from leaching tests with passive equilibrium samplers made of polyoxymethylene, as shown by Enell et al. (2016) was used. Data from these measurements (using means of $n = 3$ measurements over a 9 month-period for metal(loid)s) was then extrapolated to a 100-year timeframe using the liquid to solid ratio (L/S-ratio) and assuming that the leaching of substances from the soils will remain steady in time.

Table 1
Modeling of the main activities in scenarios S1, S2 and S3.

Process	Modeling
Electricity mix	The Swedish electricity mix was replaced in several activities with the electricity mix in the region where the WM facility is located (SE4-southern Sweden). The environmental impacts of the electricity supply mix in the region were estimated using the average approach and based on data for 2018, as shown by Papageorgiou et al. (2020).
Pyrolysis	The upstream impacts from the manufacturing of the pyrolysis unit and the transportation of the unit from the manufacturing plant to the site were modelled based on the Ecoinvent activities "Furnace production, wood chips, with silo, 5000 kW, CH" and "transport, freight, lorry >32 metric ton, EURO5, RER". The air emissions from the combustion of syngas were extracted from Sørmo et al. (2020).
Biofuel use by trucks and machinery	The transportation of materials by trucks using biodiesel was modelled based on the Ecoinvent activity "transport, freight, lorry 28 metric ton, vegetable oil methyl ester 100%, CH". A skid-steer loader and machinery using biodiesel were modelled based on the Ecoinvent activities "excavation, skid-steer loader, RER" and "diesel, burned in building machine, GLO" replacing the diesel input with biodiesel according to their heating values.
Biochar carbon sequestration.	The biochar C sequestration was of 2.4 t CO_2-eq/t biochar, assuming 80% (Azzi et al., 2019) stability and 81% carbon content (Sørmo et al., 2020).
Heat substitution	22,212 GJ of heat is required to compensate for the lower heat production in S2 and S3 (S1: 58,218 GJ - S2, S3: 36,005 GJ). Heat substitution was modelled adapting the Ecoinvent activity "heat production, softwood chips from forest, at furnace 5000 kW, state-of-the-art 2014, CH" to reflect local conditions and heat production from forest residues. The modeling of the woodchips supply chain was based on data from Hammar et al. (2015).
Backfill material production	Backfill material was modelled by the Ecoinvent activity "gravel production, crushed, CH" and adapted to match the electricity mix in SE4.

For the landfilled soil, the L/S-ratio was estimated using the method described by Birgisdóttir et al. (2007), according to which the 100-year period is divided into four time periods that reflect the changing conditions during the active and passive phase of a landfill (Table S.2.1 in S.M). For the collected leachate during the active phase of the landfill, it was assumed that 40% of the substances is routed to the effluent that is released to the environment (Öresund strait). The remaining 60% is routed to the sludge that is sent for treatment. The system boundary was set before the sludge treatment. For the reused remediated soil, the L/S-ratio was derived assuming that the average infiltration of water is 50% of the annual precipitation. It was decided to use a generic average value for the infiltration of water in the remediated soil, as its final application is unknown for the moment. Moreover, it was assumed that the lifetime of the final application of the remediated soil is 100 years, the thickness of application layer is 1 m and 100% of the leachate from the soil is emitted to the surroundings (Table S.2.1 in S.M). The average annual precipitation was estimated 758 mm based on local precipitation data from the Swedish Meteorological and Hydrological Institute (SMHI, 2020).

2.4.1. SFA sensitivity analysis

A sensitivity analysis was conducted in order to assess the influence on the results of the assumption about the infiltration of water in the reused remediated soil. The flows of the substances in S2 and S3 were estimated for 20% and 80% infiltration and compared with the original scenarios (50%).

3. Results and discussion

3.1. Mass and energy balances

The mass and energy balances from the application of MEFA are shown in Fig. 2. The mass balances show that the consumption of fossil fuels in S1 and S2 is approximately 14 and 22 times higher than in S3. The increased consumption of diesel in S1 and S2 is due to transportation of contaminated soil and backfill material, and consumption of diesel for transporting remediated soil for reuse in S2. Moreover, the consumption of biodiesel in S1 is more than 6 times higher than in S2 and S3 because of the transportation of wood waste from the WM facility to the incinerator by trucks running on biodiesel. Another finding is that in S1 and S2, virgin material is used for backfilling, something that is avoided in S3 thanks to on-site soil remediation. The energy balances indicate that the supply of heat for district heating from the wood waste incineration in S1 is 1.6 times higher than the supply of heat from pyrolysis in S2 and S3. They also reveal that a considerable amount of electricity, equal to almost 40% of the heat exported for district heating, is used for operating the pyrolysis plant.

These findings indicate that on-site remediation with biochar (S3) can provide significant fuel and virgin material savings compared to “dig and dump” (S1). By contrast, off-site remediation (S2) cannot provide fossil fuel or virgin material savings compared to S1, as it involves more transportation and requires virgin soil materials for backfilling. However, off-site remediation can reduce biodiesel consumption. Moreover, the energy balances show that there is an energy penalty in the biochar scenarios S2 and S3 due to the reduced heat production from syngas combustion and the increased consumption of auxiliary electricity in the pyrolysis plant. Hence, without input from the LCA, it is very difficult to deduce which of the three systems is more beneficial from an environmental perspective. Nevertheless, when comparing off-site with on-site biochar remediation, the latter enables more efficient use of resources.

3.2. Life cycle environmental impacts

S2 and S3 perform better than S1 in 10 out of 12 environmental impact categories, with S3 having lower impacts than S2 in all categories (Fig. 3). Notably, S2 and S3 have negative scores for Climate change, which means that, in these scenarios, greenhouse gas (GHG) uptake is larger than emissions to the atmosphere. The main reason is carbon sequestration in the biochar, which is 2.3 and 4.5 times larger than direct greenhouse gas emissions from the systems in S2 and S3, respectively. S2 and S3 perform worse than S1 only in Ionizing radiation and Fossils. The higher scores in these categories are explained by the larger electricity consumption for pyrolysis, as a percentage of electricity is from nuclear power. Due to the lower heat production from the pyrolysis plant, the consumption of woodchips is increased in scenario S2 and S3 (heat substitution). This increased consumption of biotic resources is not reflected in the Land use impact category, as woodchips come from forest residues not contributing to increased land use (but more intensive land use, for which the loss of forest carbon stocks was included).

In S1, wood waste incineration and transportation of materials are the two main contributors to most impact categories (Fig. 3). Transportation is the main contributor to Climate change, Ozone layer depletion and Fossils due to consumption of fossil fuels by trucks. It is also a significant contributor to Freshwater and terrestrial acidification, Freshwater eutrophication, Marine eutrophication, Terrestrial eutrophication, Land use, and Minerals and metals, mainly due to consumption of biodiesel. The use of biodiesel by machinery in the landfill is also the main reason why the disposal of contaminated soil contributes considerably to Freshwater Eutrophication and Land use, although land occupation by the landfill also contributes to Land use. For Minerals and metals, apart from transportation, the production of backfill material is also a hotspot.

In S2 and S3, the main environmental hotspots are pyrolysis of wood waste, heat substitution and transportation of materials (Fig. 3). Shredding and sieving contributes considerably to Freshwater and terrestrial acidification, Freshwater eutrophication, Marine eutrophication and Terrestrial eutrophication due to consumption of biodiesel. The specificity of S2 is that the excavated area is backfilled with virgin material (gravel), and that the remediated soil is re-used elsewhere, substituting the production and transport of virgin material. The impacts of these two processes roughly compensate for each other, e.g. in Minerals and metals. In S3, biochar is used on site and avoids the need for any additional backfill material, thus saving fuels for transportation.

A comparison of the environmental impacts estimated in this study with those of previous assessments of remediation techniques is generally difficult due to different choices of system boundaries and impact assessment methods. In addition, this study combined the treatment of two different waste streams. Nevertheless, a finding that is consistent with what has been reported in previous studies (Amponsah et al., 2018; Busset et al., 2012; Choi et al., 2016; Hector et al., 2012; Suer and Andersson-Sköld, 2011) is that material transportation in off-site techniques can be a major contributor to environmental impacts, especially climate change. It is should be noted, though, that for off-site remediation techniques, and waste management in general, transportation assumptions are usually context-dependent and a large source of variability between the results of different studies (Amponsah et al., 2018; Suer and Andersson-Sköld, 2011).

3.2.1. Results of the LCA sensitivity analysis

The results of the sensitivity analysis are presented in Table 1 and visualized in S.3 in S.M. The scenarios where no biodiesel is used (b-scenarios) perform worse in climate change, as the GHG emissions increase (S1b) or the emission savings decrease (S2b and S3b). There is also an increase in Ionizing radiation, Ozone layer depletion and Fossils, especially for S1b. However, there is a decrease for the b-scenarios in the other eight impact categories. Especially, for S1, where more biodiesel is used, the impacts to Freshwater eutrophication, Marine eutrophication and Land use are considerably reduced. Nevertheless, the ranking of the scenarios remained mostly unchanged. These findings indicate that there are trade-offs between the use of diesel and biodiesel. Biodiesel can provide GHG emission savings, but also causes more

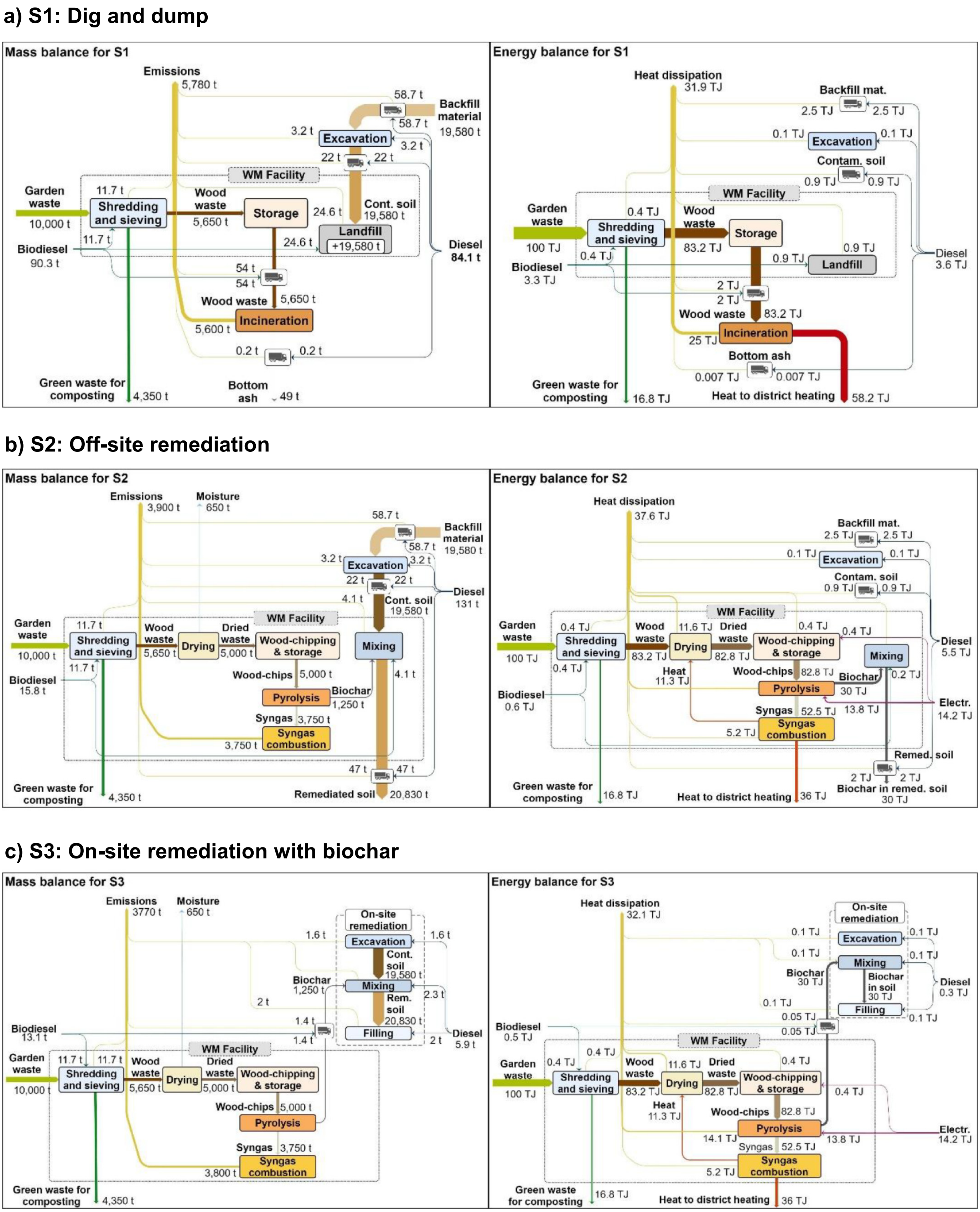

Fig. 2. Mass and energy balances for the three scenarios.

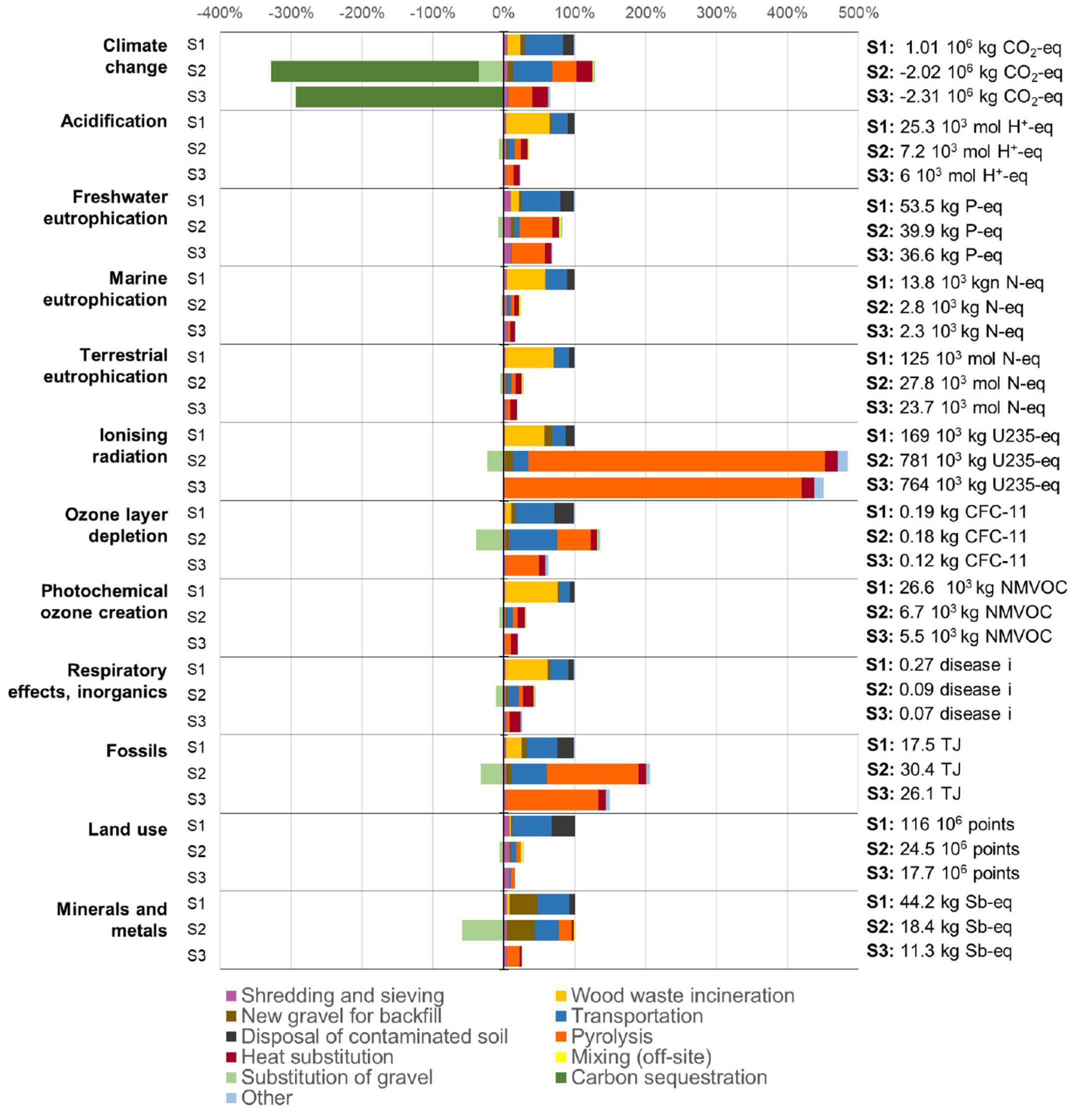

Fig. 3. Environmental impacts of S2 and S3, normalized to S1 (S1 = 100%) with process contributions. Net total values for S1-S3 are listed to the right.

impacts than diesel to several other impact categories. However, there is a large selection of raw materials for biodiesel with wide ranges of associated impacts (Cherubini and Strømman, 2011), and a choice to model another type of biodiesel could produce different findings.

In the scenarios where fossil electricity is used instead of the electricity mix in SE4 (c-scenarios), significant changes are noticed, especially in S2c and S3c. In these scenarios, even though the net climate change impact remains negative thanks to carbon sequestration in the biochar, the GHG emission savings are lower, as fossil electricity is now used in the pyrolysis plant. The use of fossil electricity is also the main reason why the contribution of S2c and S3c to Ozone layer depletion and Fossils increases considerably. In contrast, Ionizing radiation impact now decreases, as the consumed electricity does not come from nuclear power. In the other impact categories, minor or moderate changes are noticed for S2c and S3c. For S1, the most notable changes are the increase in Climate change and the decrease to Ionizing radiation. Despite the changes, the ranking of the scenarios changes only in Ionizing radiation and Ozone layer depletion.

3.3. SFA results

The L/S ratio for the landfilled contaminated soil after 100 years of leaching was estimated at 15.5 L/kg and for the remediated soil 29.2 L/kg. The estimated amount of leachate collected for treatment in S1 during the same period is 1.47 10^5 m^3 and the amount of leachate emitted to the surroundings 1.56 10^5 m^3. For S2 and S3, the estimated amount of leachate emitted to the surroundings is 6.07 10^5 m^3.

The results of the SFA for metal(loid)s are presented in Fig. 4 (the numerical values can be found in section S.4 in S.M). With the exception of Ba and Mo, the metal(loid)s emissions from the landfilled soil (S1) and the remediated soil (S2 and S3) are much lower than the sum of emissions from other processes in the lifecycle of these scenarios. For the metal(loid)s in the contaminated soil that exist at levels higher than the guideline values set by the Swedish Environmental Protection Agency (SEPA) (Ba, Cu, Pb, Hg and Zn), only a small proportion of their initial content leaches; less than 0.4% for all substances, except Ba in S2 and S3 where 1.1% leaches out. This is probably because the biochar used in the field experiments contained readily soluble Ba. Other metal(loid)s, which existed in the soil in natural concentrations, or in background concentrations to be expected in an urban environment, leach to the same degree (less than 0.8%) except Mo where 4.7% of the initial content leaches out in S1 and 25% in S2 and S3. When comparing the scenarios, smaller amounts leach from the remediated soil (S2 & S3) for Cu, Hg, Ni and Zn, and larger amounts for As, Ba, Cd, Cr, Co, Mo, Pb and V.

For PAH, the results of the SFA are more consistent (Fig. 5). The emissions of PAH from soils are several orders of magnitude higher than the

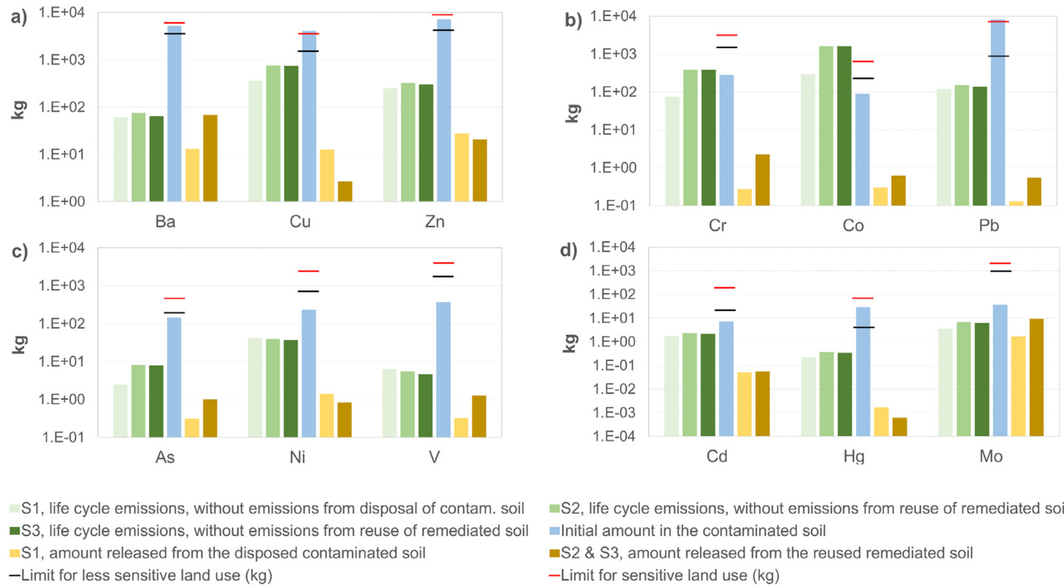

Fig. 4. Results of the SFA for metal(loid)s (in logarithmic scale and grouped to improve visual). The figure also displays the Swedish guideline values for contaminated soil for sensitive or less sensitive land uses scaled to the functional unit of the study (i.e. 19,583 kg of contaminated soil with a density of 1600 kg/m^3).

sum of PAH-emissions from other processes in the life cycle of the scenarios, with the exception of benzo(*a*)pyrene. However, only a small amount of the initial PAH content in the soil leaches out during the set timeframe of 100 years of infiltration and leaching, for all scenarios. The remediation of the contaminated soil with biochar reduced the leaching of PAH to minimal levels and, as a result, more than 99.9% of the initial content of PAH will remain in the remediated soil after the 100-year period.

Figs. 4 and 5 also display the guideline values for contaminated soil defined by SEPA (SEPA, 2009). The guideline values have been developed for two different types of land use, sensitive land use (SL) and less sensitive land use (LSL). Soils contaminated with metal(loid)s and PAHs at levels higher than the LSL values are often landfilled, while soil with contaminants at levels in between the two guideline values can be used for less sensitive land uses (e.g. industrial areas), and soil with contaminants at levels lower than SL value can be used for sensitive land uses (e.g. residential areas). The contaminated soil investigated in this study contained Cu, Pb and PAH at levels above the LSL value and thus, without treatment, it would require landfilling. The remediation with biochar seems to have the potential to stabilize PAH and certain metal(loid)s. Nevertheless, under the current legislative framework in Sweden, it is unclear whether remediation with biochar is considered as a treatment option that would enable the reuse of contaminated soil in sensitive or less sensitive land uses. For the moment, individual assessments are required for each case, based on site-specific circumstances (Flyhammar et al., 2020).

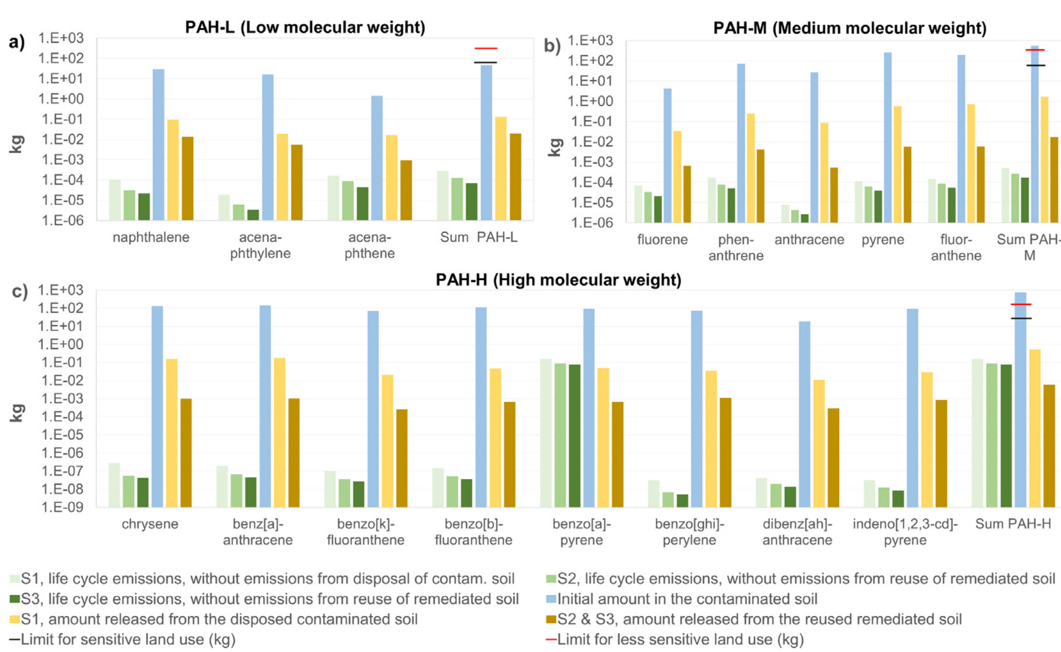

Fig. 5. Results of the SFA for PAH (in logarithmic scale). The figure also displays the Swedish guideline values for contaminated soil for sensitive or less sensitive land uses scaled to the functional unit of the study (i.e. 19,583 kg of contaminated soil with a density of 1600 kg/m^3).

3.3.1. Results of the SFA sensitivity analysis

The results of the sensitivity analysis are presented in Fig. 6. For PAH, the amounts emitted from the remediated soil after 100 years are much lower than what is emitted from the landfilled soil, regardless of the assumption on the percentage of water infiltration. However, for metal(loid)s, the results are more varied. For Cu, Hg and Ni, the leached amounts from the remediated soil are always lower than the leached amounts from the landfilled soil, regardless of the degree of infiltration, while for As, Ba, Cr, Mo, Pb and V, the leached amounts from the remediated soil are always greater. For Cd, Co and Zn, the degree of infiltration determines whether the leached amount from the remediated soil is greater than that from landfilled soil. It should be noted that for Zn, the leached amount from the remediated soil is slightly higher than the one from the landfilled soil at 80% infiltration.

3.4. Methodological reflections

3.4.1. Assumptions on biochar stability, electricity mix

The net climate change impact of the biochar scenarios (S2 and S3) was much lower than that of S1. The biochar scenarios even reached a net negative climate change impact of more than 2000 t CO_2 yr^{-1} (Table 2). This is due to a combination of assumptions and Swedish-specific circumstances, mainly: (i) the assumed biochar stability, (ii) the type of electricity used for pyrolysis, and (iii) the type of fuel used to compensate for the lower heat production from pyrolysis.

Biochar carbon sequestration is an important aspect because it contributes predominantly to the net climate change score of the studied systems, while being also an inherently uncertain term. Here, we assumed that 80% of the carbon initially present in biochar would remain in the soil for 100 years, in line with the literature (see Section 2.3.1.1). However, in addition to the inherent uncertainty of biochar stability, the characteristics of the biochar that NSR will produce are still unknown, as well as the fate and end-of-life of the remediated soil.

The pyrolysis plant consumed electricity to heat the biomass and produce biochar (3.1 MWh_e/t biochar). Globally, electricity is a driver of fossil fuel consumption and a large contributor to global GHG emissions. In Sweden, however, the average electricity emission factor is low (here estimated 100 g CO_2-eq kWh_e^{-1} in SE4) due to a high share of nuclear and hydro power. As a result, the higher consumption of electricity in the biochar scenarios was not a major contributor to the climate change impact. However, the sensitivity analysis (see Section 3.2.1) showed that if fossil electricity were used in the pyrolysis plant, the net negative climate change impacts in S2 and S3 would be reduced considerably.

Finally, we assumed the increased demand for biomass to be met by harvesting of residues from Swedish forests. Even when including emissions due to loss of forest carbon stocks because of harvesting of residues, the climate change impact of forest residues is on the lower-end of fuels commonly used for heating. Therefore, the lower bioenergy production in the biochar scenarios was not penalized in terms of climate change impact thanks to the current availability of large amounts of forest residues in Sweden.

These assumptions are decisive for the biochar systems to deliver effective climate change mitigation and make the results of this analysis specific to Sweden. The sensitivity analyses showed how the results can be adapted to other contexts.

3.4.2. Application of SFA instead of including toxicity impacts

Instead of including toxicity impacts in the LCA, the SFA was performed. This was made for three reasons. First, the final use of the remediated soil is still unknown, as it faces legal, economic and technical constraints. Therefore, the fate of the contaminants in the environment cannot be modelled accurately. Second, current LCIA methods face known limitations when it comes to estimating the toxicity of metal(loid)s relative to other substances (Lemming et al., 2010b; Nuss and Eckelman, 2014; Rosenbaum et al., 2018). In the USEtox model,

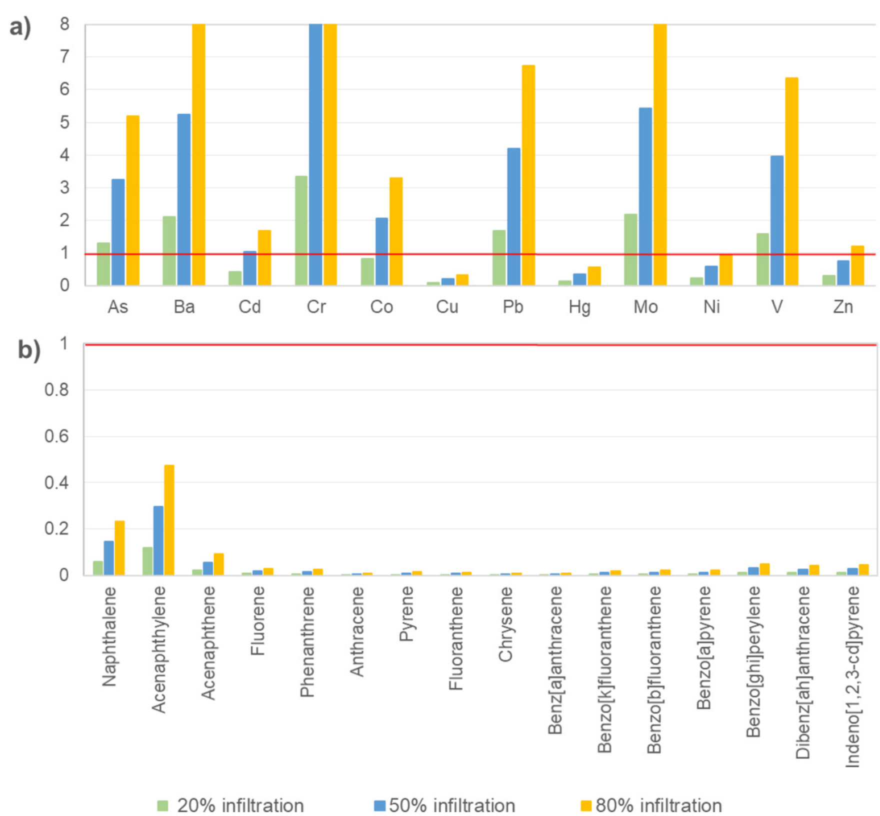

Fig. 6. The ratio of the amount of a substance leached from the remediated soil in S2 and S3 to the amount of the substance leached from the landfilled soil in S1, for different percentage of water infiltration in the remediated soil (a: metal(loid)s, b: PAH). Bars above the red line indicate a higher release to the environment of a given substance in the biochar scenarios than in the landfilling scenario.

Table 2

Results of the sensitivity analysis (S1a, S2a, S3a: initial scenarios; S1b, S2b, S3b: scenarios with no biodiesel use; S1c, S2c, S3c: scenarios with fossil electricity). For b- and c-scenarios, the percentage change ($\frac{\text{Initial value}-\text{Initial value}}{|\text{Initial value}|} \cdot 100$) in relation to the initial scenarios is given.

		Climate change (10^6 kg CO_2-eq)	Freshwater and terrestrial acidification (10^3 mol H + -eq)	Freshwater eutrophication (kg P-eq)	Marine eutrophication (10^3 kg N-eq)	Terrestrial eutrophication (10^3 kg N-eq)	Ionizing radiation (10^3 kg U235-eq)	Ozone layer depletion (kg CFC-11)	Photochemical ozone creation (10^3 kg NMVOC)	Respiratory effects, inorganics (Disease incidences)	Fossils (TJ)	Land use (10^6 points)	Minerals and metals (kg Sb-eq)
Initial scenarios	S1a	1.01	25.3	53.5	13.8	125	169	0.192	26.6	0.270	17.5	116	44.2
	S2a	−2.02	7.20	39.9	2.78	27.8	781	0.184	6.67	0.0885	30.4	24.5	18.4
	S3a	−2.31	5.96	36.6	2.34	23.7	764	0.120	5.54	0.0706	26.1	17.7	11.3
No biofuel	S1b	9.7%	−18%	−68%	−34%	−18%	7.6%	25%	−5.0%	−5.4%	15%	−73%	−12%
	S2b	0.92%	−8.3%	−14%	−23%	−10%	0.31%	4.6%	−0.44%	3.2%	1.5%	−54%	−4.6%
	S3b	0.63%	−9.4%	−12%	−24%	−11%	0.25%	5.7%	−2.4%	−0.43%	1.4%	−61%	−6.0%
Fossil electricity	S1c	27%	0.093%	−6.0%	0.52%	0.55%	−69%	16%	0.84%	−0.34%	13%	−0.65%	−0.41%
	S2c	89%	3.8%	−51%	18%	17%	−96%	110%	24%	−4.4%	49%	−18%	−2.1%
	S3c	78%	4.5%	−56%	21%	20%	−98%	170%	28%	−5.5%	57%	−25%	−3.5%

which is used in the ILCD 2.0 impact assessment method, the characterization factors for metal(loid)s are considered to be interim, meaning that impact assessment results must be interpreted carefully if dominated by metal(loid)s (Fantke et al., 2017). This is partly due to processes like metal(loid)s complexation not yet taken into account in toxicity compartment models. Finally, aggregation of contaminant emissions at the impact level would hide differences in treatment efficiency between contaminants.

3.5. Research limitations

The pyrolysis of wood waste in the WM site was modelled based on data from the literature (Sørmo et al., 2020), as at the time of the study the pyrolysis unit was not operational. Hence, to examine the validity of our findings, emission tests in the new plant at the NSR site are recommended.

The lysimeter data, on which the SFA is based, is from three sampling occasions during a 9 month-period and only represent one type of contaminated soil and a specific type of biochar. The tested biochar seems to be suitable for binding mainly PAH and cations, like Cu, Hg, Ni and Zn, something that is in agreement with what has been reported in the literature (Beesley et al., 2011; Cheng et al., 2020; Shen et al., 2016; Zama et al., 2018). Nevertheless, for the three cations, Pb, Cd and Co, the sorption by biochar was lower than reported in the literature (Hilber et al., 2017; Medyńska-Juraszek et al., 2020), which is likely to be due to their already low concentrations in the studied soil pore water and/or different properties of the biochar. For substances that exist as anions and/or are pH and redox sensitive, such as As, Cr, Mo and V, the tested biochar seems not to be suitable for stabilization. The low sorption effectiveness of biochar for As and Mo is in accordance with what has been reported in the literature (Hilber et al., 2017), but for Cr and V good sorption potential has also been reported, depending though on the type of feedstock (Thomas et al., 2020; Yu et al., 2020). Therefore the results of the SFA depend on the type of biochar used and the type of contaminated soil.

Moreover, the calculations in the SFA are based on the assumption that the leaching from the soils is constant over the entire time period, which is a rough approximation. In fact, the leaching can both decrease and increase over time, due to changed soil chemical conditions. It should also be noted that the type of application will have a large effect on the amount of water that infiltrates through the reused remediated soil and thus the leaching of certain substances. It is also unclear what the fate of substances that accumulate in the soils will be, especially in the long-term, and how environmental conditions may influence the bioavailability of contaminants in soil.

The abovementioned issues highlight that the SFA results are site specific and dependent on the soil quality, the type of biochar, the type of application and the bioavailability of contaminants in the soil. Thus, generalized conclusions on the potential risks to ecosystem quality and human health cannot be drawn at this stage. A comprehensive risk assessment of future biochar uses for remediation of contaminated soils using data from site-specific applications could be a next step.

4. Conclusions

In this study, LCA was coupled with MEFA and SFA in order to assess the environmental impacts of biochar systems for remediation of contaminated soil and energy recovery from wood waste, and compare them with the impacts of the conventional “dig and dump” technique By combining these methods, a more detailed description and assessment of the investigated systems could be performed, while avoiding some of the limitations in toxicity impacts assessment in LCA.

The MEFA indicated that on-site remediation with biochar can provide considerable fuel and virgin material savings compared to “dig and dump” and off-site remediation. However, the pyrolysis of wood waste for biochar production generates less heat than incineration and requires a fair amount of auxiliary electricity. The LCA showed that both on-site and off-site remediation with biochar performed better than “dig and dump” in 10 of the 12 environmental impact categories, with on-site remediation performing better, as it saved fuel and backfill material. Both on-site and off-site remediation showed large reductions in climate change impact compared with the “dig and dump” technique, thanks to high biochar carbon sequestration and specificities of the Swedish energy system. The two biochar systems showed increased impacts only in two environmental impact categories, mainly due to increased electricity consumption in pyrolysis. They also resulted in increased biomass demand to maintain energy production. The SFA indicated that leaching of PAH from the remediated soil was lower than the landfilled soil, regardless of water infiltration level. For metal(loid)s, no straightforward conclusion could be made due to high sensitivity of the results to water infiltration. Therefore, the reuse of biochar-remediated soils in urban environment requires further evaluation, taking into account site-specific information, like contaminant concentration in the treated soils, background concentration levels at each site, and variability in runoff reaching the site.

Overall, in Sweden's current context, the use of biochar for remediation of contaminated soils seems to be an environmentally sound alternative to the “dig and dump” technique. Still, safe applications for the biochar-remediated soils need to be further developed and tested in specific cases, and assessed from risk and regulatory perspectives.

CRediT authorship contribution statement

Asterios Papageorgiou: Conceptualization, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft, Writing – review & editing. **Elias S. Azzi:** Conceptualization, Methodology, Validation, Visualization, Writing – original draft, Writing – review & editing. **Anja Enell:** Conceptualization, Writing – review & editing, Project administration, Funding acquisition. **Cecilia Sundberg:** Conceptualization, Methodology, Writing – review & editing, Supervision, Funding acquisition.

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 at https://doi.org/10.1016/j.scitotenv.2021.145953.

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