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Alexandre Tisserant

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Department of Energy and Process Engineering

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Preface

The thesis has been submitted to the Faculty of Engineering Science in partial fulfillment of the degree of Philosophiae Doctor. This work was carried out at the Industrial Ecology Programme and the Department of Energy and Process Engineering at the Norwegian University of Science and Technology in Trondheim, Norway, in the period 2018-2022.

Alexandre Tisserant

Trondheim, August 2022

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Abstract

In 2015 the international community signed the Paris agreement with the aim of maintaining global warming below 2°C. Negative emissions technologies (NETs) that are able of removing atmospheric carbon dioxide for storage are key component of climate scenarios that reach the Paris agreement. Those technologies may be required to accompany transition to a zero emissions society, to offset diffuse and hard to decarbonized greenhouse gas emissions and potentially help bring back down the temperature if the climate target is overshot. Several NETs have been proposed, some concerns exist regarding their efficacy, the amount of material, energy or land they may require and how they may negatively impact other sustainable goals, such as biodiversity and food production. Investing the climate mitigation potential of these technologies requires to account for emissions happening along their supply chains and quantifying co-benefits and/or trade-offs against other sustainable or environmental targets. This thesis takes this perspective to investigate a specific NET: biochar. Biochar emerged in the recent years as a win–win option that can act both as a carbon sink and as amendment improving soil quality, increase fertility, and water holding capacity.

The first part of the work was to develop a framework for analysis of biochar systems by reviewing literature on biochar, its global warming mitigation potential and how it may affect climate both locally and globally, its soil effects, the potential use of its co-products (e.g. syngas and bio-oil) and previous work regarding its environmental assessment (Chapter 1). Biochar can supply substantial negative emissions at the global level, while its co-products can be used for energy production offsetting fossil emissions, while condensing bio-oil and storing it in geological deposits could provide additional carbon sequestration. Biochar can improve agronomic performance and reduce some soil emissions, but also increase others. Those effects vary depending on its production condition, soil conditions and background climate, and field management of biochar. Biochar can also affect local climate by changing land surface energy balance and the emissions of some short-lived gases that have a climate effect.

The rest of the work was to quantify negative emission potentials and associated co-benefits and trade-offs of large-scale deployment of biochar in Norway, using forest residues (Chapter 2) and in Europe, using both crop and forest residues (Chapter 3). Three system are investigated biochar, biochar and energy production in a combined heat and power cycle (CHP), and sequestration of both biochar and bio-oil. Biochar can offset about 1.3% (17.4%) of Norwegian (agricultural) GHG emissions per year, and between 1.6% and 3.7% (14.2% and 31.7%) of European (agricultural) emissions per year depending on feedstock supply availability. Sequestration of the bio-oil can almost double those potentials. Additional climate mitigation from the CHP are larger in Europe due to more carbon-intensive energy mix compared to Norway. Biochar can increase crop yield, soil water retention over large areas in Europe, and reduce agricultural impacts in some environmental categories, but increase in others. However, when taking the life-cycle perspective biochar systems are typically net positive impacts generator due to emissions happening in the supply-chain.

The sooner net zero emissions can be achieved, the less reliant on deploying NETs we will be to reach the Paris agreement. Biochar production is a well-known process, is cost effective and can ramp up negative emissions while improving soil health before other NET deployment maturity.

List of publications

Tisserant, A. & Cherubini, F. (2019) Potentials, Limitations, Co-Benefits, and Trade-Offs of Biochar Applications to Soils for Climate Change Mitigation, *Land*, 8:12

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Author contribution: Research co-design, literature review and writing

Tisserant, A., Morales, M., Cavalett, O., O'Toole, A., Weldon, S., Rasse, D. & Cherubini, F. (2022) Life-cycle assessment to unravel co-benefits and trade-offs of large-scale biochar deployment in Norwegian agriculture. *Resources, Conservation and Recycling*, 179

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Author contribution: Research co-design, data collection, modeling, visualization, analysis and writing

Tisserant, A., Hu, X., Liu, Q., Xie, Z., Zhao, W. & Cherubini, F, Biochar and its potential to deliver negative emissions and better soil quality in Europe

Submitted to *Earth's Future*

Author contribution: Research co-design, data collection, modeling, visualization, analysis and writing

Chapter 1. Introduction

At the time of writing this thesis, the earth is experiencing a global increase in temperature of about 1 °C since pre-industrial time[1]. However, mankind is already experiencing 1.5 °C[2], as land surfaces tend to warm faster than the ocean. Most of the past 40 years have been warmer than 1971-2000 global temperature average (Figure 1). Global warming represents a threat for food security, water availability, livelihood, economic development, and biodiversity[3].

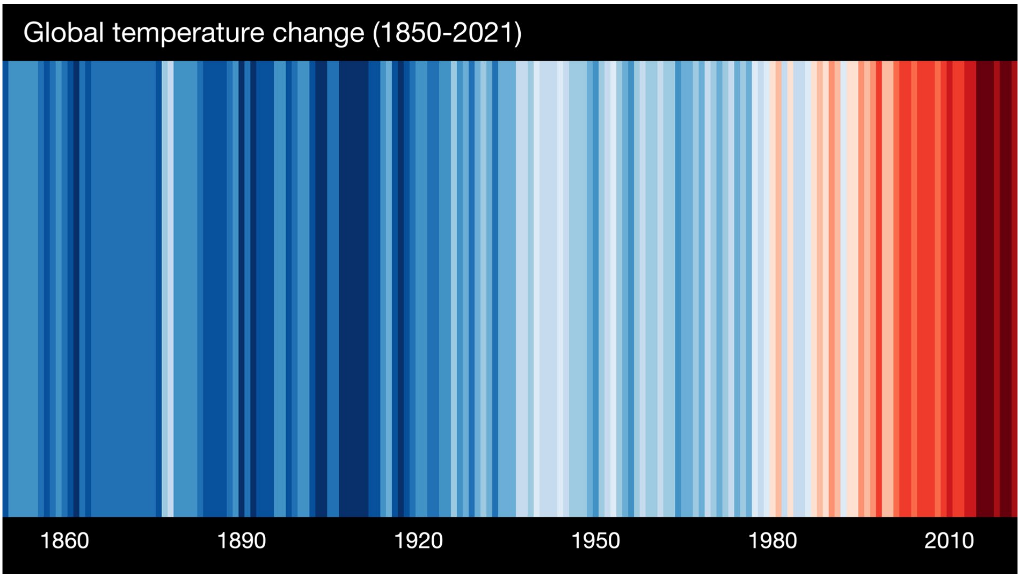

Figure 1: Global temperature change over the period 1850-2021 (each line represents one year) compared to average temperature over 1971-2000. Blue lines represent cooler year compared to the 1971-2000 period, red lines show warmer years. Intensity in color represents the size of the gap between a given year and the 1971-2000 average. Source: Prof. Ed Hawkins (University of Reading), https://showyourstripes.info/s

In this context, the United Nations agreed on limiting risks and impacts of climate change by signing the Paris Agreement in 2015[4]. The agreement states the objective of limiting global temperature increase to 2 °C while pursuing efforts in limiting to 1.5 °C. The Intergovernmental Panel on Climate Change (IPCC) analyzed climate mitigation pathways and concludes that reaching the Paris agreement requires to achieve net zero CO_2 emissions by early 2050 to stay below 1.5 °C warming or early 2070 to limit temperature increase to 2 °C[5]. These pathways typically rely on deep decarbonization of the economy by switching to low carbon energy production such as renewable energies, implementing carbon capture and storage (CCS), reducing emissions of other GHG particularly methane and nitrous oxides, improving energy efficiencies and reducing losses[5].

Most of the pathways achieving the Paris agreement also rely on dedicated technologies capable of capturing back atmospheric carbon[5]. Termed as negative emissions technologies (NETs), they aim at compensating for diffuse emissions such as methane, agricultural emissions, in hard to decarbonize sectors (such as maritime and air transport) and for delays in deploying low carbon technologies. Deployment of NETs can also allow for net negative emissions, effectively decreasing atmospheric CO_2 concentration to allow for slow cooling of the Earth. It is also clear that the sooner deep cuts in GHG emissions happen across the economy, the less reliant on NETs the pathways are[5].

Several NETs, also sometimes referred to as carbon dioxide removal (CDR) technologies, are currently being suggested. NETs aim to remove atmospheric carbon dioxide by (1) increasing natural sinks for carbon; or (2) using engineering methods to capture the CO_2[6]. Proposed NETs are both land and ocean based, most of them relies on enhanced photosynthesis and two are based on chemical capture of CO_2. They consist of afforestation and reforestation, bioenergy with carbon capture and storage (BECCS), biochar, enhanced weathering, soil carbon sequestration, ocean fertilization and direct air capture (DACCS)[6]. Climate change is however only on of the current global challenges, where biodiversity, food security, fair livelihood and inequalities, air and water pollution are also topics of increasing importance. It is therefore required to develop solutions that can deliver across several sustainable issues, or at least do add pressure on other issues[5].

Many climate mitigation options and fossil fuel alternatives, such as bioenergy, biomaterial and some NETs, typically rely on land biomass. These technologies are therefore likely to compete for resources, while increasing land use for biomass production will compete with other important sustainable development goals, particularly food production and biodiversity, while increased inputs uses such as irrigation, fertilizers and pesticides may be unsustainable and affect air and water quality[7].

Increasing land competition, climate change and soil degradation processes put stress on food security and affordability[8,9]. There is therefore a need to fight climate change while not competing with land use, maintain or improve food production and help rehabilitate degraded soils. Over the last decades, biochar has gain traction as suitable carbon sequestration method that can be produced from readily available low-value by-products, and has interesting agronomic properties that can help rehabilitate soil. The present thesis focuses on the environmental and sustainable analysis of biochar.

1.1 Biochar at the interface of climate mitigation and agriculture

Biochar is the solid remainder of a process called pyrolysis, where biomass is brought up at high temperature in the absence of oxygen. Atmospheric carbon is captured during biomass growth and converted into a stable carbon structure during pyrolysis that can be stored in soils for centuries[10,11], and as such is considered a NET. Biochar can be produced from available forest and crop residues that are not currently used[12], improving the material efficiency of current forest and crop production, without requiring additional land, irrigation or inputs, such as fertilizers or pesticides. Biochar production also produces a liquid phase, bio-oil (i.e. a mixture of water and organic compounds), and gaseous phase, syngas[13]. Bio-oil and syngas can be used for energy recovery for example by upgrading bio-oil to liquid biofuel or burnt to recover heat feeding a combined heat and power unit, potentially offsetting carbon emissions as well. Bio-oil is also rich in carbon and could be directly stored in geological deposits, increasing the carbon sequestration potential of pyrolysis process[13,14].

Applied to agricultural soils, biochar is advocated to have beneficial effects on plants and soils. Biochar can increase crop yield and enhance food production[15,16]. Positive effects are mostly observed in acidic, weathered soils in the tropics, while positive yield response in fertile land is more limited. Other positive plant physiology responses are also typically observed such as increase photosynthesis[17], better root system[18] and water use efficiency[19]. Soil quality may also be improved via increased native soil carbon stock[20,21], improved soil aggregation[22], water infiltration and retention[23]. Soil health can also be improved with increased biomass and microbial activities leading to higher nutrient cycling and ecosystem stability of agricultural soil[24]. Biochar has also been studied as a soil remediation tool for its capability to reduce availability of heavy metals and sorption of organic compound such as pesticides, limiting plants' intake[25].

Current agricultural practices use large amount of fertilizer to achieve high crop yield. These fertilizers, in particular nitrogen, are responsible to emissions that have a climate effects and affect air and water quality, acidification, or the stratospheric ozone layer[26,27]. Biochar tend to reduce soil emissions of N_2O and NO_x and of nitrogen leaching, but may increase volatilization of NH_3[28,29].

Effects of biochar in soils are usually beneficial and could allow to co-deliver on other environmental impacts and sustainable goals, such as reduced nitrogen pollution, improved soil quality and crop yields. However, those effects are highly dependent on local weather and soil conditions, type of biochar feedstock and pyrolysis conditions, while emissions along biochar's supply chain could balance out any savings in soil effects.

1.2 Quantifying climate mitigation, co-benefits, and trade-offs potentials of biochar

As any other technology, biochar mobilizes a supply chain that requires energy and materials leading to emissions to air, water, and soil. At the same time, different emissions can affect one or several environmental aspects and/or several emissions affect a single environmental aspect but at varying degrees. Both aspects are relevant to assess net potential of biochar deployment across various environmental impacts. For example, how much emissions happening in biochar's supply chain can offset its carbon sequestration potential? Or what is the climate mitigation potential of reduced soil N_2O emissions compared to its carbon sequestration?

Life-cycle assessment (LCA) is a standardized method that allows to address those two aspects. It allows assess the environmental impacts of a product and identify potential trade-offs between one or more environmental impacts[30]. In a first stage, inventories of emissions (e.g. kg CO_2 emitted) and resources used (e.g. kg of iron ore used) across the different processes involved in the supply chain are collected. Then, impact assessment methods are used to weights the various emissions relevant to a given environmental category, such as global warming or marine eutrophication.

LCA focuses on environmental impacts and does not include indicators related to agronomic performance, soil quality or soil health. Biochar will typically have different effect depending on the type of crop grown, local climate, local soil conditions (e.g. soil pH, texture), the amount of biochar applied etc. However recent modeling approaches allow to link crop yield and soil response to biochar application taking into account those local conditions[21,28,31,32]. These modeling approaches can be used to identify potential agronomic co-benefits or trade-offs to the application of biochar using knowledge on local soil and climate conditions for example.

1.3 Thesis contribution

Biochar has been studied at various scale from laboratory testing to field trials, generating data on its effects on soil and plants under different conditions, and have been summarized in various meta-analysis[33]. However, most environmental analysis of biochar only consider its climate mitigation potential[34,35]. Studies focus on large-scale climate mitigation potential of biochar considering local, regional or global feedstock availability [36–40]. They typically consider only the carbon sequestration potential of biochar but does not include other soil effects or emissions along the supply chain. On the other hand, many LCA studies of biochar systems have been performed that account for supply-chain emissions[41–50], but inconsistently represent biochar's effects on soil and do not scale up their results to available resources to provide large-scale climate mitigation.

At the same time, most studies only quantify climate mitigation potential of biochar of its carbon sequestration and of GHGs, but supply chains also generate emissions of short-lived gases that can have strong climate effects, while changes in field emissions of NO_x and NH_3 due to biochar application will also affect climate over short period of time. There is therefore a need to quantify climate effects of biochar systems using different climate metrics representative of short, medium and long-term impacts to identify whether or not there is a trade-off between short- and long-term climate mitigation.

In this thesis I aim at understanding implication of biochar deployment and identifying potential co-benefits or trade-offs between climate mitigation and other sustainable goals, such as food production, soil quality, air and water quality. In a second step, the goal is to quantify those effects at different level a variety of soil effects of biochar considering local soil conditions, accounting for emissions happening in the supply chain, and constraining biochar deployment to yearly available biomass resources that do not compete with land use. Different application levels of biochar are also considered as they affect both level of response of soils to biochar, but also constrain the amount of land that can be treated yearly at a given biomass supply potential. This level of details has not been achieved thus far in large-scale assessment of biochar deployment and could provide interesting insights on where most benefits can be achieved and where biochar may induce strong trade-offs.

Chapter two provide a qualitative review of its global warming mitigation potential, its potential effect on global and local climates, its agronomic benefits in terms of crop yield, soil quality and soil emissions. Identifying key controlling factors on those aspects regarding local climate and soil conditions, biochar's feedstock and management options (e.g. application rate of biochar). It also reviews current environmental analysis of biochar systems that use LCA. Chapter three quantifies the climate mitigation, and environmental co-benefits or trade-offs in terms of air and water quality, stratospheric ozone depletion of biochar deployment in Norway using forest residues. Different uses of biochar's co-products are also investigated, where only biochar is produced, energy recovery from co-products provide heat and electricity, or the bio-oil is also recovered for carbon sequestration in fossil fuel geological deposits. Chapter four explores the same biochar production options and analysis as chapter three but extended to Europe. Spatially explicit data on locally available forest and crop residues is used to derive the amount of biochar that can be produced and the amount of cropland that can be treated each year. It also investigates in more detail agronomic effects providing spatially explicit maps of co-benefits or trade-offs in terms of crop yields and improved soil water retention. It also considers two application rates of biochar (i.e. either 5 or 30 t ha^{-1}), as soil response to biochar depends on its amount applied. Finally, chapter five summarizes the main findings, limitations and discusses the policy relevance of the work and concludes on potential future work.

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Chapter 2. Potentials, Limitations, Co-Benefits, and Trade-Offs of Biochar Applications to Soils for Climate Change Mitigation

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Review

Potentials, Limitations, Co-Benefits, and Trade-Offs of Biochar Applications to Soils for Climate Change Mitigation

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Abstract: Biochar is one of the most affordable negative emission technologies (NET) at hand for future large-scale deployment of carbon dioxide removal (CDR), which is typically found essential to stabilizing global temperature rise at relatively low levels. Biochar has also attracted attention as a soil amendment capable of improving yield and soil quality and of reducing soil greenhouse gas (GHG) emissions. In this work, we review the literature on biochar production potential and its effects on climate, food security, ecosystems, and toxicity. We identify three key factors that are largely affecting the environmental performance of biochar application to agricultural soils: (1) production condition during pyrolysis, (2) soil conditions and background climate, and (3) field management of biochar. Biochar production using only forest or crop residues can achieve up to 10% of the required CDR for 1.5 °C pathways and about 25% for 2 °C pathways; the consideration of dedicated crops as biochar feedstocks increases the CDR potential up to 15–35% and 35–50%, respectively. A quantitative review of life-cycle assessment (LCA) studies of biochar systems shows that the total climate change assessment of biochar ranges between a net emission of 0.04 tCO_2eq and a net reduction of 1.67 tCO_2eq per tonnes feedstock. The wide range of values is due to different assumptions in the LCA studies, such as type of feedstock, biochar stability in soils, soil emissions, substitution effects, and methodological issues. Potential trade-offs between climate mitigation and other environmental impact categories include particulate matter, acidification, and eutrophication and mostly depend on the background energy system considered and on whether residues or dedicated feedstocks are used for biochar production. Overall, our review finds that biochar in soils presents relatively low risks in terms of negative environmental impacts and can improve soil quality and that decisions regarding feedstock mix and pyrolysis conditions can be optimized to maximize climate benefits and to reduce trade-offs under different soil conditions. However, more knowledge on the fate of biochar in freshwater systems and as black carbon emissions is required, as they represent potential negative consequences for climate and toxicity. Biochar systems also interact with the climate through many complex mechanisms (i.e., surface albedo, black carbon emissions from soils, etc.) or with water bodies through leaching of nutrients. These effects are complex and the lack of simplified metrics and approaches prevents their routine inclusion in environmental assessment studies. Specific emission factors produced from more sophisticated climate and ecosystem models are instrumental to increasing the resolution and accuracy of environmental sustainability analysis of biochar systems and can ultimately improve the characterization of the heterogeneities of varying local conditions and combinations of type feedstock, conversion process, soil conditions, and application practice.

Keywords: biochar; pyrolysis; food security; climate mitigation; negative emission technology; carbon dioxide removal; life-cycle assessment; environmental assessment; biogeochemical cycles; biophysical effects

1. Introduction

Human activities affect the earth system globally, pushing on some planet boundaries [1]. Crossing the boundary for climate change is a major concern as global climate feedbacks could push toward a "Hothouse Earth" pathway and could deeply affect the biosphere and society globally [2]. For this reason, the Paris agreement states the goal of "Holding the increase in the global average temperature to well below 2 °C above preindustrial levels and pursuing efforts to limit the temperature increase to 1.5 °C" [3].

Reaching the Paris agreement is challenging as some economic sectors are hard to decarbonize (e.g., transportation) or are subject to lock-in (e.g., lifetime of power plants). Capture of carbon from the atmosphere or at the plant will be required to offset unavoidable emissions. Inertia in transforming the economic system will likely lead to an overshoot of the Paris target, and atmospheric capture of carbon will be required during a period of net negative emissions to lower back temperature by 2100. Even under scenario pathways of lifestyle changes and faster deployment of renewable electrification, net negative emissions are required [4].

Several negative emission technologies (NET) for carbon dioxide removal (CDR) are discussed in the literature: afforestation and reforestation, bioenergy with carbon capture and storage (BECCS), biochar, enhanced weathering, soil carbon sequestration, ocean fertilization, and direct air capture (DACCS) [5]. Several of them raise concerns regarding energy requirements, land competition, toxicity, and potential unexplored long-term consequences [5–8].

Biochar [9] emerged in the recent years as a win–win option that can act both as a carbon sink and as an amendment improving soil quality, increase fertility, and water holding capacity, thereby preventing risks for land degradation [10]. Production of biochar occurs via a thermochemical process called pyrolysis. Pyrolysis is the thermal decomposition of biomass at high temperature and in the absence of or under very low oxygen concentration and is associated with the production of three by-products: biochar (solid), bio-oil (liquid), and syngas (gas).

Biochar production and application to agricultural soils interact with the environment and climate system in multiple complex ways. There are emissions from feedstock collection, transport, and biochar production via pyrolysis, and biochar applications affect soil emission balance (e.g., N_2O, CH_4, NH_3, and NO_x). Biochar also influences the climate system by complex mechanisms, including the long-term storage of biogenic carbon in soils and changing soil reflectivity (e.g., albedo) by darkening the surface. All these aspects are relevant for environmental assessment of climate change, eutrophication, acidification, and human health, for example.

There are many existing review studies on biochar, but studies that include this variety of factors in an integrated framework are rare or only includes some of them [11–15]. An increasing number of studies assesses the role of biochar as a negative emissions technology, with a quantification of technical, economic, and sustainable large-scale deployment potential [5,14,16,17]. Individual studies focus on different aspects of its production systems and on one or more environmental implication(s), such as the long-term stability and effect on soil organic carbon (SOC) [18–21], its effect on soil physical and hydraulic properties [22–25], soil degradation [26,27], agricultural yield [28–32], greenhouse gas balance (GHG) [33–44], nitrogen availability and emissions [37–40,45–47], that of phosphorus [48–50], biochar's toxicity [51–53], remediation potential [54–59], and effects on pesticides [60–62]. These review studies generally show that the type of biomass feedstock, biochar production conditions, local soil properties, and management decisions all modulate soil and environmental responses after biochar production and application and that undesired outcomes in terms of environmental effects and/or agricultural yield are possible under certain conditions [13,31,63].

In this review, we provide an assessment of the state of the art of biochar systems for soil amendment, taking a life-cycle perspective by studying the relevant environmental aspects of its production and usage from feedstock provision, pyrolysis and long-term application to soils, and potential substitution effects. We discuss the variety of effects that biochar can have on the climate system (e.g., carbon sequestration and changes in surface albedo and in soil GHG emissions), its potential for climate change mitigation, the connections with food security, and other environmental concerns such as toxicity and ecosystems quality. We identify three main aspects that control the environmental performance of biochar systems: (1) biomass feedstock type and biochar production conditions, (2) soil properties and local climate conditions, and (3) biochar management and application practices. These three aspects are used as the main factors to explain variability of biochar effects on environmental and climate systems in the different sections of this review.

This review is structured as follows. First, we provide an overview of the various aspects of biochar deployment along its supply chain and use phase that are relevant for (1) climate mitigation and adaptation, (2) food security and soil quality, and (3) toxicity and ecosystems resilience. Global potential estimates of carbon sequestration from large-scale biochar deployment are presented and discussed in relation to the needs of CDR for specific temperature stabilization targets. We then discuss appropriate biochar feedstocks for carbon sequestration and agronomic purposes, followed by an overview of biochar production systems in terms of pyrolysis technologies and possible uses of by-products. Climate regulations mechanisms affected by biochar application to soils related to biogeochemical (e.g., global carbon, nitrogen, and water cycles) and biogeophysical (e.g., albedo and evapotranspiration) aspects are also reviewed. The different possible combinations in terms of biomass feedstocks, biochar production processes, local soil type and climate conditions, and agricultural management practice are discussed as key factors explaining variability in environmental outcomes of biochar systems. Interactions among those aspects and with factors outside the biochar value chain are also discussed, and life-cycle assessment (LCA) studies of biochar systems are reviewed. Before concluding, we discuss aspects relevant to consider for social and ethical implications of biochar deployment.

2. Biochar and Climate Change Mitigation

There are different aspects through which biochar systems interact with the climate. These include storage of carbon in soils, GHG emissions from the biochar value chain, changes in surface albedo from biochar application to agricultural soils, etc. These aspects are complex and highly case-specific. This section introduces these aspects and their interactions; we refer the reader to the next sections for more details on controlling factors and scale of effect. Figure 1 summarizes how biochar interacts with the climate system once incorporated in the field (not all mechanisms may happen in all cases, and some mechanisms can result in either cooling or warming depending on local conditions).

In terms of GHG emissions, biochar aims at mitigating climate change by capturing and storing atmospheric carbon in recalcitrant form, while the combined effect of increased soil organic carbon (SOC) stability and biomass yield after biochar application may also lead to an increase in stock of soil carbon in agroecosystems. Collection and transport of biomass residues require energy and is associated with GHG emissions. At the same time, collection of residues will avoid GHG emissions due to their decomposition at the cost of potential losses of SOC (see Section 6). Combustion of pyrolysis gas leads to emissions of CO_2, SO_2, NO_x, and N_2O. When pyrolysis gas is not burned, pyrolysis exhausts will be composed of CH_4, CO and non-methane volatile organic carbon (NMVOCs). Soil gas balances are also affected by biochar application (N_2O, CH_4, NO_x, and NH_3), and potentially increase emissions of black carbon and soil dust aerosols. Especially N_2O and CH_4 are powerful GHGs. Near-term climate forcers (NTCF) NO_x, NH_3, CO, and NMVOCs are aerosol precursors that affect climate in different ways depending on the emitting region [64]. Black carbon is an aerosol with strong warming potential (average 900 kgCO$_2$eq/kg black carbon) [65], and potential emissions from field need to be better assessed. Black carbon and particulate matter can also be emitted during pyrolysis, particularly under conversion processes based on low-technology conditions [66].

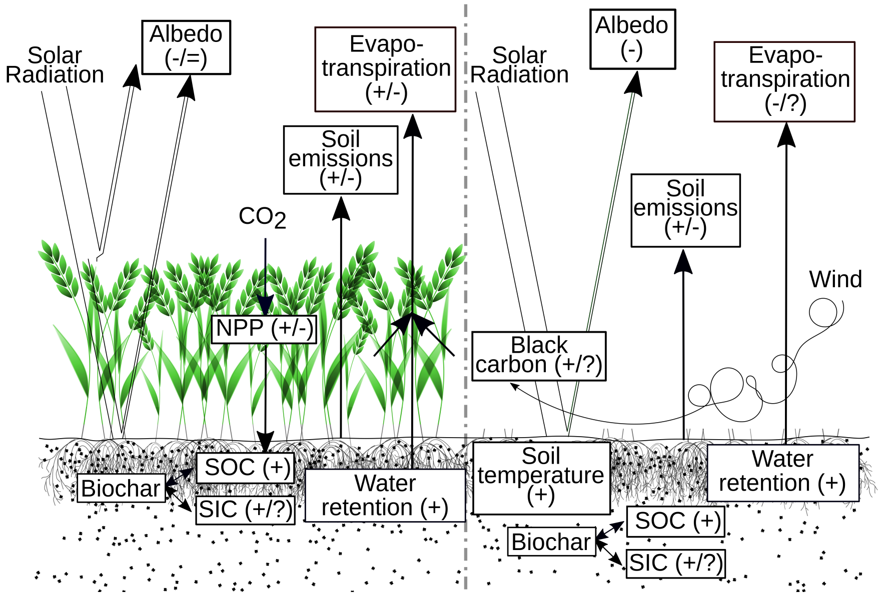

Figure 1. Biochar's effects on climate under cultivated field (**left**) or fallow (**right**) conditions. Signs in parenthesis indicate biochar's effect on the variable compared to control without biochar: (+) increased, (−) decreased, (=) unchanged, (?) there is limited data available for assessment. We refer the reader to Sections 2 and 8 for more detailed descriptions of the different mechanisms of how biochar in soil may affect the climate system. ***Soil Organic Carbon (SOC)***: Biochar has a positive direct effect on SOC by providing recalcitrant carbon and an indirect positive effect on SOC by stabilization of soil carbon. Some biochar carbon may be leached from soils or transported by wind (see Section 8.1) ***Soil Inorganic Carbon (SIC):*** Biochar's effect on SIC is still limited in scientific evidence, but a preliminary study shows that biochar increases SIC stock both directly and indirectly. ***Albedo***: Biochar tends to make soils darker and, hence, to reduce surface albedo. However, the presence of a vegetation canopy or snow cover can dampen these effects. ***Soil emissions***: Changes in soil emissions depend on the gas (i.e., N_2O, CH_4, NO_x, and NH_3), biochar properties, and soil conditions (see Sections 8.1 and 8.2 for more details). ***Water retention***: Biochar increases soil water retention and plant available water, making more water available for evapotranspiration under cultivation and evaporation under fallow. ***Evapotranspiration***: Under cultivation, biochar has a contrasting effect on evapotranspiration depending on soil condition and climate (e.g., precipitation level and energy limitation for evapotranspiration) and can increase or decrease plant water use efficiency. Under fallow, biochar tends to reduce evaporation; however, more evidence is needed. ***Net Primary Productivity (NPP)***: Biochar has mixed effects on NPP depending on soil conditions; increased NPP fixes more carbon in vegetation, increasing residues left on field and root and increasing root exudates, which may participate in increasing SOC (see Section 8.1). ***Black Carbon***: During application of biochar, tilling operation microparticles of black carbon can be transported by wind. ***Soil temperature***: In the absence of crop canopy, soil temperature increases and daily soil temperature fluctuations, which can affect sensible heat flux, water evaporation, and SOC degradation rate. Under cultivation, biochar tends to decrease soil temperature fluctuations.

The Earth's surface also influences climate. Incoming energy at the soil surface is balanced by upwelling emissions of long-wave radiations and sensible, latent, and ground heat fluxes [67]. Reduced albedo due to biochar application increases short-wave absorption, making more solar energy available at the surface. Changes in soil albedo after biochar application is estimated to reduce its climate benefit by about 13–30% [68,69]. Albedo's effect on climate depends on the amount of incoming radiation [70]. The warming effect due to reduction in soil albedo will be lower at higher latitudes, while biochar's effects on aerosol and soil moisture may affect cloud formation and the amount of radiation reaching the soil. Under cold climates and snow condition, potential transport and deposition of biochar's black carbon will decrease snow and ice albedo, and biochar-amended snow-free patches may also increase the snow melting rate on field.

Reduction in soil albedo, changes in soil water availability, and soil thermal properties due to biochar will control the proportion between sensible, latent, and ground heat fluxes, thus controlling surface temperature. Genesio and colleagues measured changes in albedo and modeled the surface energy balance of a durum wheat field in Italy [71]. They found that, overall, biochar increases all energy fluxes at a seasonal and yearly scale, while it increases soil temperatures during bare soil regime [71]. Increased soil moisture can help mitigate drought. It also increases total evapotranspiration potential, which has a cooling effect, and thus can help mitigate heat waves. Finally, soil moisture is positively correlated to the level of precipitation [72]. Biochar's effect on soil water retention and plant water availability may represent an interesting adaptation to climate change. Most of these biochar–climate interactions are discussed in detail in the following sections.

3. Biochar, Food Security, and Soil Quality

Biochar's positive effect on agricultural yield is often cited as an important co-benefit of its carbon sequestration. However, some negative yield responses are also observed. Biochar's effect on plants physiology and soil contaminants can also indirectly impair food security. Whether biochar increases or decreases risk of soil degradation or help reclaim degraded soil are also important aspects for food security [73,74].

Several meta-analyses have investigated biochar's effect on agricultural yield [28,29,75,76]. Highly weathered soils that are acidic with low cation exchange capacity (CEC) and receive little agricultural inputs, as found in tropical regions, see a positive response to biochar application in terms of yield [29]. An average increase in yield of 25% is observed in tropical soils, while biochar has no or very little positive or even negative effects in temperate soil [29]. In tropical soils, high nutrient biochars (e.g., from manure) have a stronger positive effect on yield [29]. Increased soil moisture can increase yield in temperate regions that have less weathered soils and higher agricultural inputs [77,78]. Negative yield responses are mostly observed under alkaline soil conditions [29] (potentially limiting P supply to plants [79]). Application rates larger than 50 t biochar/ha in temperate soils lead to statistically significant negative effects on yields, while tropical soil see their yield responses increase at application rates between 50–150 t biochar/ha [29] (see Table S1 in Supplementary Information (SI) based on References [29,80]).

Plant physiological responses to biochar are not all well understood yet [81]. Concerns exist regarding the role of biochar in reducing plant defense [82,83]. Biochar may lead to improved or reduced plant response toward foliar and soilborn pathogens [84]. In addition, biochar can immobilize pesticides [60], which may reduce efficiency of treatments against soilborne pathogens. Similarly, better growing conditions for crops may increase weed competition [31], while herbicides are made less efficient [85]. Biochar may affect early development of crops, as the albedo effect may warm soils and ease germination, while sorbed volatile organic carbons (VOCs) on biochar and free radical generation may impair germination [86,87].

Agricultural soil erosion is the most serious threat to agriculture sustainability and food security [88,89]. A decrease in agricultural yield on highly eroded fields can be as high as 65–80% [90]. Biochar can help preventing soil erosion but can also have negative effects [22,23]. Better soil hydrology and soil wet aggregate stability reduce water run-off and soil loss, but tillage can cancel biochar's positive effect [23]. Biochar may have no effect or negative effects on soil dry aggregate stability, potentially increasing wind erosion risks [22].

Biochar also has other potential positive effects on soils. Saline and sodic soils occupy 500 Mha of land and is expected to increase under climate change [91]. Biochar improves plant response and alleviates stress on crops grown under drought and salt stress [92,93], but its ash content may increase salinization risks and stress [93].

Removal of farm products and application of nitrogen fertilizers are responsible for acidification of croplands [74,94], which leads to lower nutrient availability, toxicity issues, nutrient leaching, and soil emissions. Biochar can act as a liming agent and can increase soil buffering capacity through carbonates, increased cation exchange capacity (CEC), and base cation provision [95,96].

As a drawback, biochar may be a source of contamination by bringing polyaromatic hydrocarbons (PAHs), dioxins, VOCs , and heavy metals depending on its feedstock and production conditions [51,53,97]. However, biochar can immobilize heavy metals through sorption, precipitation, and pH/oxidoreduction reactions and was shown to consistently reduce plant heavy metals concentration [59]. Biochar may act as a source of PAHs in soils; however, observed concentrations remain below the maximum acceptable limit [51]. Biochar has the capacity to adsorb pesticides, potentially leading to accumulation in soils [60,62], though they become less available and may also be degraded by biochar [60,98].

4. Biochar, Toxicity, and Ecosystems

Emissions of reactive nitrogen from combustion and N fertilizer use have repercussions on human and ecosystem health via a variety of pathways [99]. These nitrogen emissions have effects on soil acidification [100], toxicity and human health [101], global nutrient biogeochemical cycle, and deposition [102] with implications for ocean and land carbon sinks [103] and land ecosystems [104]. Gas emissions during pyrolysis [66,105,106] and dust and black carbon emissions from soils are also potential threats to human and ecosystem health [107].

Liu and colleagues [45] investigated global biochar deployment scenarios aiming at maximizing plant production, minimizing soil N_2O emissions or soil total N losses, e.g., N_2O, NH_3, and N leaching (while limiting other negative outcomes). Because each N species fate in soil is affected differently by biochar, they find that, depending on the scenario considered, NH_3 volatilization decreases by 12% or increases by 29%. Once in the atmosphere, NH_3 creates particulate matter, playing a role for atmospheric haze (with cooling effect), that has serious effect on human health and alters the climate system. Part of NH_3 is deposited back on terrestrial and aquatic ecosystems, where it causes acidification, eutrophication, and fertilization [99].

Biochar affects soil NO_x emissions while, during pyrolysis, part of the nitrogen in feedstock is volatilized and lead to emissions of NO_x precursors. Biochar effect on soil NO_x emissions is, however, much less studied then N_2O or NH_3 emissions. NO_x increases tropospheric ozone concentrations, causing particulate matter (PM). Both ozone and PM have serious effects on human health; ozone can be also deposited and can reduce productivity of ecosystem (including crops). NO_x also causes acidification, eutrophication, and fertilization once deposited on terrestrial or aquatic ecosystems [99]. Pourhashem and colleagues estimates the value of reducing soil NO_x emissions due to biochar application to be 660 million dollars per year in the US [101].

Biochar reduces nitrate (NO_3^-) leaching but has mixed effect on leaching of ammonium (NH_4^+) and phosphorus [23]. Nitrogen and phosphorus leaching from agricultural fields are well known for eutrophication of freshwater and marine ecosystem [108] and also affect groundwater and drinking water quality. In their global assessment, Liu and colleagues [45] estimate that biochar has potential to reduce soil N leaching by 12–29% (see more information in the previous paragraph). In Blanco-Canqui's review [23], he reports changes in P leaching from decrease of 62% to increase by 152%.

Exposition to black carbon PM can happen during the handling of biochar, in its application to soils, and by wind transport. Grinding of biochar to increase its specific surface area for adsorption purposes can transform 2–5% of biochar to particles under 2.5 µm and 10 µm [109]. Aging, abrasion, and tilling in soils and being transported can also reduce biochar to fine particles [110–112].

Black carbon particles are toxic to humans and ecosystems. In particular, they represent a risk for lung and heart diseases and could transport contaminants from soils to humans [107].

Dissolved organic carbon (DOC) affects aquatic ecosystems through a variety of mechanisms, such as metal toxicity, turbidity, higher temperature stratification, and lower light penetration in lakes [113,114]. Biochar was found to increase leaching of dissolved organic carbon [23,115]. Leachate of soil biochar may also be toxic to terrestrial and water ecosystems due to heavy metals and PAHs [52,53]. Dissolved biochar molecules may produce toxic reactive oxygen species under sunlight in water ecosystems [116,117].

If carefully produced, biochar has limited potential to pollute soils and can help as a remediation tool [53]. As a remediation tool, biochar decreases bioaccumulation of heavy metals in plants, thus reducing risk toward human health [118,119] (see Table S2 based on References [51,62,97,120,121]). However, its sorption capacity renders pesticides less mobile and reduces their biodegradation [60,61]. This has mixed benefits as it reduces the fate of pesticides in groundwater and toxicity toward non-targeted organisms, reduces pesticide efficiency toward pests, and might require higher application rates, increasing expositions to humans and ecosystems [62,122].

5. Negative Emissions and Biochar

In terms of CDR potential, available estimates for biochar deployment largely differ in the literature and range from 0.65 to 35 $GtCO_2eq$/year depending on a wide set of assumptions [9,123–131]. For example, Lehmann and colleagues [9] provide an estimate of 2.13 $GtCO_2eq$/yr assuming a replacement of burning management in shifting agriculture to biochar production, the use of forestry and crop residues, and current bioenergy needs met by pyrolysis. They also estimate biochar potential between 20–34 $GtCO_2eq$/yr assuming that all 2100 bioenergy needs are fulfilled by pyrolysis with a yield of 30.6 kgC sequestered by GJ of bioenergy. Laird and colleagues [128] give a current potential of biochar of 1.5–3.3 $GtCO_2eq$/yr, assuming 50% and 67% of crop and aboveground forestry residues are used, respectively, while the upper bound adds 67% of belowground forest residues, thinning of disease-ridden forest, and application of pyrolysis to avoid 50% of human biomass burning emissions.

Most studies estimating negative emissions from biochar deployment constrain its production to the supply of forestry and crop residues. However, dedicated crops grown on abandoned and marginal land can provide additional feedstock for biochar production with potential co-benefits (see Section 6). Woolf and colleagues [125] assume a potential of 0.6–1.1 $GtCO_2eq$ biochar per year produced from crops grown on degraded and abandoned land. It adds 26–32% of biochar production potential on top of their estimates of 1.3–3.0 $GtCO_2eq$ biochar produced from residues. Powell and Lenton [126] include production of bioenergy crops on abandoned land in their assessment of CDR potential toward 2050. These crops are used only for BECCS in their scenarios but would represent a biochar production of about 3–6.2 $GtCO_2eq$/yr in 2050 (they assume 10 t dry matter/hectare on 0.33–0.69 Gha made available in 2050). Adding both potentials (from residues and dedicated crops), a total of 7.8–10.3 $GtCO_2eq$/yr is reached.

Schmidt and colleagues [17] investigate the potential to sequester bio-oil in geological deposits in addition to biochar. This strategy increases the carbon sequestration efficiency by a factor $\sim$1.7 (from 30–42% to 53–74%) [17], which corresponds to an increase in negative emission from 0.65–35 $GtCO_2eq$/yr (biochar only) to 1.1–60 $GtCO_2eq$/yr (biochar and bio-oil).

Figure 2 shows the range of potential estimates of negative emissions from biochar within the context of the scale and deployment rate of negative emissions that are required to stay within a given temperature warming level since preindustrial times [132]. The size of required negative emissions is related to the time of peak emissions and the size of the overshoot, with longer delays to curb emissions requiring larger deployment of NETs in the second half of the century [16,133]. Needs for CDR remain below 5 $GtCO_2eq$/yr before 2030 but rapidly increase to more than 20 $GtCO_2eq$/yr in the case of meeting the Paris agreement with high chance to overshoot the 1.5 °C target ('1.5 °C high overshoot').

With estimated negative emissions between 0.1–3.3 $GtCO_2eq/yr$, biochar can play a significant role in providing CDR for all temperature pathways until 2030, which is the year considered for current potentials (Figure 2). From 2050 onward, biochar produced from residues has a negative emission potential of 1.1–4.9 $GtCO_2eq/yr$, which is in the bottom 10% of the required range for the '1.5 °C low overshoot' pathways. Including biochar production from dedicated crops at 7.8–10 $GtCO_2eq/yr$ [126], biochar is respectively in the 35% and 15% ranges of CDR deployment for pathways meeting the 1.5 °C target with low or high overshoot toward 2100 and in the 50–35% range of the required deployment for a lower 2 °C pathway (see Table S3 in SI). If bio-oil is also sequestered under the most optimistic biochar deployment scenario (∼17.5 $GtCO_2eq/yr$), biochar sequestration provides substantial CDR for pathways consistent with the 1.5 °C target (see Table S3 in SI).

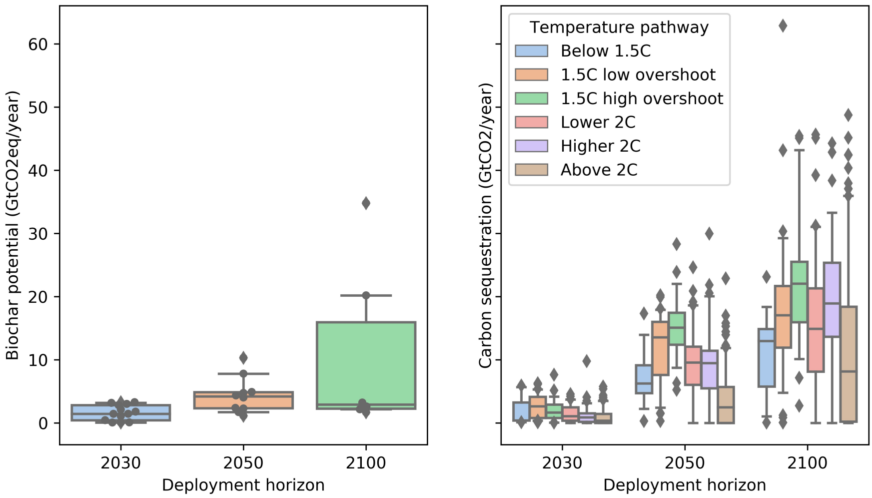

Figure 2. Biochar's potential for carbon dioxide removal (CDR) at different time horizons (**left**) and negative emission requirements for different time horizons and temperature pathways (**right**): Estimates of biochar potential come from 10 studies [9,123–131]. Studies estimating the current biochar's potential are under the 2030 horizon. CDR requirement numbers are taken from the Integrated Assessment Modeling Consortium 1.5 °C scenario explorer [132]. CDR requirements are calculated as the sum of the variables "Carbon Sequestration | CCS", "Carbon Sequestration | Land Use", "Carbon Sequestration | Direct Air Capture", and "Carbon Sequestration | Enhanced Weathering". Only 9 pathways are consistent with 'below 1.5 °C', therefore the range of CDR must be taken carefully. Boxes represents the 1st and 3rd quartiles, with the middle line being the median; whiskers represent the 5th and 95th percentiles. Black diamonds represent outliers, i.e., values below the 5th percentile or above the 95th percentile, while black circles represent individual biochar production potential estimates.

The use of dedicated energy crops is essential to achieving large negative emissions from biochar deployment. In order to avoid competition for land and detrimental effects for food security and natural systems, changes in existing land management and higher efficiency of land use is needed to free areas for the sustainable growth of dedicated biomass crops for biochar production. This can be achieved via parallel developments of multiple response options aiming at reduction of food waste, dietary changes (lower meat consumption and declines in pasture lands), and increase in yields [134].

The global land sink for biochar application is estimated between 2 Gha and up to the total agricultural land area of 4.5 Gha [9,125,131,135]. Estimates for carbon sink can be as high as 2200 $GtCO_2eq$; however, these estimates either assume that all agricultural areas are receive biochar [135] or at high application rates (≥170 t biochar/ha) [131]. These estimates seem unrealistic given that it would include all grassland and that significant negative yield responses are observed in temperate soils at application rates over 50 t biochar/ha (see Section 3). Woolf and colleagues [125] consider that about 2 Gha of global agricultural land (16% of global land area) is available to receive biochar (1.5 Gha of global cropland and 20% of 2.5 Gha of global pasture). The biochar application rate is commonly between 20–50 t biochar per hectare and could be as high as 150 t biochar per hectare, in certain regions [29] (see Section 3). Total biochar sink potential could be somewhere between 180–410 $GtCO_2eq$ considering a global average application rate between 30–70 t biochar per hectare (at 80% carbon in biochar [17]). It represents only 5–15% of the range of global cumulative CDR needed (see Table S4 in SI based on Reference [132]).

In addition to the biochar contribution to CDR deployment, biochar can provide further co-benefits, such as yield improvements, bioenergy, and reduction in soil GHG emissions, and can represent a NET that can co-deliver across multiple societal challenges. In their biochar assessment, Woolf and colleagues [125] estimated that carbon sequestration in biochar accounts for about 50% of its climate mitigation potential, while 30% comes from replacement of fossil-fuel energy by pyrolysis energy and 20% from avoided soil emissions of N_2O and CH_4. Their estimate of total carbon abatement from biochar is between 3.7–6.6 $GtCO_2eq$ per year (see also Section 9).

In general, potential production and carbon storage of biochar is much lower than BECCS but biochar is more technologically mature, can be deployed at lower costs, and has multiple co-benefits [5,136]. Biochar thus represents a practicable solution to ramp up negative emissions in the short-medium term, before large-scale implementation of BECCS will become feasible.

6. Feedstock Types and Supply

Biochar can be produced from a variety of feedstocks, which influence its composition and effects on soils. Carbon-rich feedstocks such as lignocellulosic biomass allow for higher carbon content in biochar. In particular, woody materials contain less extractives (e.g., sugars and metabolites) and more lignin than leafy, herbaceous materials and, therefore, are richer in carbon [137–140]. Higher levels of lignin are also associated with higher levels of aromatization and larger aromatic clusters that are key indicators of biochar recalcitrance and stability in soils [141–143].

On the other hand, organic waste products such as manure, sewage sludge, or food wastes contain less carbon [144] but are richer in nutrients (N, P, and K) [144] and, therefore, are more attractive for agronomic purposes than lignocellulosic biomass. However, biochar made from organic waste products are more alkaline than lignocellulosic biomass [145,146] and have higher salt content, which may also lead to negative effects for plant growth and soil biota due to increases in soil pH and salinity [147–149]. There are also higher risks that biochar can contain toxic compounds (e.g., polychlorinated dibenzo-p-dioxins and -furans—PCDD/Fs) formed during pyrolysis [120,150] with potential toxic effects on soils. The higher heavy metal concentration in sewage sludge and digestate can represent an additional risk of soil contamination [53,97,150].

There are different patterns for the supply of biochar feedstock that is associated with positive and negative side effects. Forest and crop residues are usually the by-products of other production systems and are usually associated with little additional upstream negative environmental impacts. However, their availability is limited and there can be competition for their use, as they can also serve other purposes (e.g., energy production or animal feed). Trade-offs between different uses will lead to trade-offs or co-benefits between different environmental concerns. Using manures (or digestate) could allow to recycle nutrients to agricultural land (see Section 8.2), while avoiding emissions (e.g., CO_2, N_2O, CH_4, NO_x, and NH_3) [151,152] associated with their handling (i.e., composting or land spreading) that have negative effects, for example, on climate, acidification, and human health. Collection of forestry or crop residues can have consequences for soil carbon [153,154], at least for the short-term; for nutrient cycling; and for maintaining soil integrity (e.g., soil structure, soil biodiversity, and erosion prevention) [154,155], but at the same time, it prevents emissions from organic matter decomposition (e.g., CO_2 and N_2O) [156]. The use of dedicated crops can achieve the largest supply potential for biochar production, but it can lead to competition for land, food security, biodiversity, and environmental degradation from fertilizer and pesticide use [6,7]. On the other hand, bioenergy crops grown on marginal land or abandoned cropland are often win–win solutions in terms of renewable energy supply, increases in soil organic carbon, and a variety of ecosystem services relative to annual crops [157–159].

7. Biochar Production

Pyrolysis consists of a large family of processes and reactor technologies. Heating rates and temperature characterize different types of pyrolysis, which yield different mixtures of the three products [160,161] (Table 1). Slow pyrolysis (i.e., low heating rate and moderate temperature) favors biochar formation, fast pyrolysis, bio-oil, gasification, and syngas. With increasing pyrolysis temperature, biochar yield decreases but carbon content and aromatic condensation of biochar increases, suggesting a trade-off between higher recalcitrance at the expense of biochar yields [143,145,161,162].

Table 1. Summary of typical operating conditions and product yield of the main pyrolysis processes [163–166].

	Slow Pyrolysis	Fast Pyrolysis	Gasification
Pyrolysis temperature (°C)	250–750	550–1000	$\geq$500
Heating rate (°C/s)	0.1–1	10–200	5–100
Feedstock particle size (mm)	5–50	$\leq$1	0.2–10
Solid residence time	450–550 s up to days	0.5–10 s	$\geq$1 h
Vapor residence time	5–30 min	$\sim$1 s	10–20 s
Biochar yield (%)	45–20	5–30	$\sim$5
Bio-oil yield (%)	40–50	50–75	$\sim$10
Syngas yield (%)	10–25	5–35	$\sim$85

Fast pyrolysis and gasification are more suited for energy recovery purposes, as they favor production of bio-oil or syngas, respectively [160,161]. Biochar produced under these conditions are less suited for climate change mitigation and application to soils due to lower yield and carbon content (low carbon storage efficiency), energy costs during pretreatment of feedstock and pyrolysis, and higher risks of contamination (e.g., PAHs, dioxins, and VOCs) [17,51,53,120,143,144,167].

Both biochar's feedstock and pyrolysis conditions influence the biochar's properties. Table S5 (based on References [96,144,145,168–181]) in the supplementary information shows a summary of key biochar properties together with indications of how feedstock selection and pyrolysis conditions influence them.

Multiple uses are possible for the biochar coproducts. Bio-oil can be used directly for energy and heat purposes, but its high oxygen content, high water content (typically between 20–30%), and low pH represent a challenge for its direct use [139,165]. Refining and upgrading of bio-oil is required for its use as bio-fuel but at the expense of increased costs and decreased energy efficiency [139,165]. Bio-oil is investigated as an additional option to provide carbon sequestration. Long-term storage of carbon can be achieved by pumping bio-oil directly in geological formation [17]. Biorefining of bio-oil has also potential to replace some petroleum-based feedstocks in the chemical industry [182] for the production of asphalt paving substitution, slow release fertilizer, pesticides and wood preservatives, resins and adhesives, or carbon fiber. Integrated in products, bio-oil would represent temporary carbon sequestration, similar to wood in wood products [183].

Syngas can be used in turbines to produce electricity or to provide heat to sustain the pyrolysis and/or to dry the feedstock [184]. Under certain conditions, syngas production during slow pyrolysis contains enough energy required to sustain the pyrolysis [185].

Biochar production is a key step for its overall environmental sustainability profile. Thermal decomposition of biomass leads to formation of a wide variety of compounds that can represent an environmental risk if not properly handled. Bio-oil and tars contain compounds toxic to humans and ecosystems [106,186]. Biochar can contain PAHs and VOCs sorbed on its surface that can be toxic to soil biota and induce physiological responses in plants, such as affecting seed germination and nutrient uptake [51,86,120]. Syngas, on the other hand, consists mostly of a mixture of CO_2, CO, CH_4, and H_2 gases that have negative effects on climate or are toxic: 5–28% of biomass nitrogen is transferred to the bio-oil or gas phase during pyrolysis and emitted as N_2O or NO_x [46]. Chlorine and sulfur can also be vaporized, leading to emissions of CH_3Cl, dioxins, and H_2S [150,187]. Appropriate handling of the biochar coproducts is required to mitigate these risks. In particular, even without energy or heat recovery, combustion of pyrolysis gases (both vaporized bio-oil and syngas) is preferred to lower emissions of toxic compounds [66,105].

8. Biochar in Soils: Biogeochemical and Biophysical Effects

Biochar affects soil physical (e.g., density, aeration, and colour), chemical (e.g., pH and oxidoreduction potential), and biological properties (e.g., macrobial biomass and community composition). These changes in soil conditions have consequences on global biogeochemical cycles (e.g., carbon, nutrient, and water cycles) and biophysical balance (e.g., soil albedo and temperature; surface energy balance) of the Earth's system. In this section, we describe how biochar interacts with the Earth's system and provide main controls of the interaction regarding biochar properties, soil type, and management.

8.1. Soil Carbon

The main process through which biochar interacts with the carbon cycle is by sequestering atmospheric CO_2 during vegetation growth and by storing a large fraction of this carbon in soils in recalcitrant form.

Biochar is made of a highly recalcitrant carbon structure toward biotic and abiotic reactions [20,188]. Estimation of residence time under field conditions range from 6 to 5448 years [189]. Biochar decomposition follows a two-pool behavior, with a labile fraction that is quickly degraded and a recalcitrant fraction respectively estimated at 3 and 97% in a meta-analysis [20]. Based on the meta-analysis [18], average decomposition rates of biochar quickly drop from 0.6433%/day to 0.0024%/day after one year, due to depletion of the labile pool and then slow degradation of the recalcitrant pool [18]. Table 2 presents key production parameters increasing biochar recalcitrance: pyrolysis time (>3 h) and temperature (>400 °C). Biochar decomposition in soils is lower under acidic conditions (steep decrease between pH 6 and 5), dry climates (<40% moisture), and lower temperature.

Table 2. Key parameters controlling biochar stability in soils.

Controlling Factors		Observations	Ref
Biochar	Pyrolysis time	Pyrolysis reaction time longer than 3 h markedly decreases decomposition rate of biochar.	[18]
	Pyrolysis temperature	Pyrolysis temperature over 400 °C produces more stable biochar.	[18,20]
	Carbon content	Higher biochar carbon content is linked to lower H/C_{org} ratio and higher degree of aromatic condensation of biochar, which are important control of its stability. Carbon content over 70% have significantly lower decomposition rates.	[18,142,171]
Soil	pH	Soils with low pH show lower degradation rates of biochar. Decomposition rate is increased by 272% from pH 5 to pH 6.	[18]
	Moisture	Increasing soil moisture increases biochar decomposition rates by 200% from 40% to 70% water content.	[18]
	Temperature	A 20 °C increase in temperature leads to a 53% increase in decomposition rate.	[18]
	C/N ratio	Biochar decomposition rate decreases with increasing soil C/N ratio but increases with soil organic carbon content. Addition of nutrient has no effect on biochar decomposition rate. It seems to indicate that biochar decomposition is more controlled by readily available C for energy than by nutrient limitations.	[20,190,191]
	Mineralogy	Higher clay content in soils lowers biochar decomposition rates. Recalcitrance of biochar is also increased by the presence of certain soil minerals that slow down its oxidation or by stabilizing dissolvable and undissolvable biochar. However, the effect of mineralogy on biochar stability is still not much investigated.	[20,190,191]

H/C_{org}: Hydrogen to organic carbon in biochar (excluding carbonates in biochar's ash); C/N: carbon to nitrogen ratio.

Biochar carbon can also be lost to CO_2 outside the field. Fresh and aged biochar can release DOC [21,192] and be leached out and transported to freshwater systems [115]. Biochar's DOC contains aliphatic, carboxyl carbons and small condensed aromatic molecules [116,192]. Small and aliphatic DOCs are labile and can be oxidized in water systems [117]. Condensed aromatic DOCs are considered more recalcitrant but are also preferentially photooxidized [117,193]. About 8–13% of this condensed-aromatic DOC may undergo complete photooxidation and may return as CO_2, while 68–91% is partially photooxidated, potentially increasing its biolability [117]. Fate of the leached DOC from biochar needs to be better assessed and quantified, as most existing studies rely on DOC from natural charcoal produced from wildfires (differing in properties [194]).

Besides being recalcitrant, biochar has also the potential to stabilize native soil organic carbon (i.e., negative priming effect on SOC), mostly due to SOC adsorption on its surface [195]. Biochar application can stimulate microbial activity after application, leading to a temporary positive priming effect but becoming negative after 2 years [19,20]. Based on a meta-analysis [19,20], time after application is the major control on biochar's priming effect. Main controls leading to negative priming include biochar production conditions: feedstock (mostly negative priming for woody and crop feedstock, mostly positive for manures and sludges), carbon content (>50%), and temperature (>500 °C). Soils poor in nutrient (C/N ratio > 11–12), with low carbon content (SOC < 1%), and acidic (pH < 6) are more likely to lead to a positive priming (see Table S6 in SI based on References [19,20,196–198]).

In addition to favoring stabilization of native SOC, biochar is also reported to increase soil microbial biomass [43,199], to improve root traits and biomass [200], to stabilize recent carbon inputs [198,201], and to increase sequestration of non-charred soil carbon [196,202,203], suggesting a positive carbon sequestration feedback of biochar addition to soils. For example, on cropland with old charcoal deposits from former charcoal-kiln sites (>120 years), concentration of non-charcoal C in soils is 1 to 1.4 times higher and contains 1.6–1.7 times more crop-derived carbon in the black spots than in adjacent soils without biochar [198,204].

The effects of biochar on soil inorganic carbon (SIC) are little explored. Dong and colleagues [205] found that biochar increases total inorganic carbon by 20–62% in the 0–20 cm soil layer and by 13–31% in the 20–40 cm soil layer [205].

Soils can be a source of methane under anoxic conditions (e.g., paddy rice) or a sink of methane, where it is oxidated in upland soils [206]. Soil methane uptake represents about the size of anthropogenic emissions from rice cultivation or from biomass and biofuel. Biochar has a mixed effect on soil methane emission and uptake (see Table S7 in SI) [33,35,36,41]. Biochar can reduce methane production rates from source soils and its uptake in sink soils [35]. Cong and colleagues [36] find that only biochar addition to coarse, upland soil with moderate (10–20 g/kg soil) amounts of SOC leads to significant increase in the methane sink capacity, while coarse paddy rice soil with low carbon may see their methane release markedly increased (though not statically significant). Overall, Ji and colleagues [35] estimated that emission reductions from paddy rice will be offset by reductions in methane uptake in soils.

During production and handling and after field application, weathering and tilling can reduce biochar to very fine particles [110–112]. Tunnel experiments suggest that biochar particles can also be transported by wind after soil incorporation [109,111,112,207]. Three out of eight soils saw its dry aggregate stability reduced by biochar [22], increasing susceptibility to wind erosion, soil dust emissions, and potentially transport of black carbon particles. High content of monovalent cation in biochar (Na^+ and K^+) may increase dust emission, while coarse and dry soil seem more responsive to biochar addition [112]. Regarding management, wet application of biochar and deep incorporation into soil can mitigate black carbon emission from soils [107]. Li and colleagues found that biochar increased PM10 black carbon emission by 4–10 times [112]. More studies, in particular, under field conditions, are needed to evaluate the potential of wind transportation of biochar particles from field and derive realistic emission factors.

8.2. N and P Cycles

Biochar affects nutrients cycling in soils via sorption and retention, increasing or decreasing their bioavailability by reducing or increasing emissions and leaching. Pyrolysis can volatilize some of the biomass's nitrogen, but about 75% remains in biochar in fixed forms, with relatively low availability [150]. Biomass's phosphorus remains mostly in the biochar, but its soil availability decreases with increasing pyrolysis temperature. Biochar has been suggested as a potential slow release P fertilizer [50,150].

On a larger scale, biochar will influence cycling of nutrients at the ecosystem level by changing leaching and deposition of nutrients and by modifying the cycling of nutrients in residues. Collection of residues will remove nutrients that would otherwise be cycled within the ecosystem (e.g., forest) or field. Depending on whether nutrient availability and losses in the field are increased, biochar application will affect how much fertilizer need may be reduced or increased, while changes in nutrient losses from the soil will affect global nutrient cycles.

Biochar influences soil cycling of nitrogen via its surface chemistry [176,208], effect on soil pH, and response of microbial communities [37,46]. Biochar application tend to decrease NO_3^- and NH_4^+ concentrations in soils by respectively 11 and 12% [48,209]. Co-application of fertilizer, particularly organic, or co-composted biochars can alleviate the risk of N supply shortage to plants [48,209,210]. From a meta-analysis, biochar beneficially increases N_2 fixation in soil by 63% and decreases N leaching by 26% [46] (see Table S8 in the SI based on References [37,48,144,209]).

Crop yield enhancement effect of biochar is linked to increased N fertilizer efficiency and plant N uptake [32,211]. In their meta-analysis on plant N uptake, Liu and colleagues [46] found that biochar significantly increase the N uptake when produced at high temperature (>500 °C) from manures, in soils with low pH (<5), and in CEC (<5 cmol/kg) and that high application rate (>80 t biochar/ha) of biochar could significantly decrease N uptake in plants. Overall, they estimated an increase in N uptake of 12%.

Increasing N fertilizer efficiency due to biochar allows to lower the application rate, indirectly reducing soil N emissions and leaching. Biochar has also a direct effect as it reduces soil emissions of N_2O by 32–38% on average after biochar application to soils [37,46]; however, its effect may only be transient, lasting up to one year after application [37]. At the field scale, biochar's effect on N_2O emissions ranges from reduction by 17% or increase by 1% [38]. Biochar decreases 47–67% soil NO_x emissions [212], though it has been less studied than N_2O emission reduction. Effect of biochar on NH_3 volatilization varies greatly, with a meta-analysis suggesting overall increase of 19% [46] and another one suggesting no net effect overall [47].

Soil N_2O emissions mitigation from biochar amendment are higher with lower biochar's H/C_{org} at higher biochar application rate and in finer textured soils. Mitigation of N_2O emissions increases also with an increased application rate of biochar and under urea and nitrate fertilization (Table 3) [37,39,40,46]. The link between soil N_2O emission mitigation and biochar's H/C_{org} ratio is made in only one meta-analysis [39], though others mention that higher temperature (generally linked to lower H/C_{org} ratios) and higher carbon content biochars lead to higher mitigation potential [37,46]. However, two meta-analyses find that mitigation of N_2O emissions by biochar may only be transient [37,38]. Verhoeven and colleagues [38] conclude that no clear factors under field conditions (e.g., biochar properties, soil conditions, or management) control N_2O mitigation and suggest that some previous observations may be an artifact of compiling nonindependent experiments. Lower biochar application rates, generally lower moisture content, a lack of homogeneous biochar incorporation, and overall less controlled conditions are reasons why less reductions are observed under field conditions compared to incubation studies [38,39]. Some conclusions among meta-analysis are contradicting, for example, the effect of soil pH (References [37,40] vs. References [46]) or of soil organic matter (Reference [37] vs. Reference [46]).

Reduction in soil NO_x emissions is more pronounced in acidic soils and with straw biochar compared to manure biochar and under all type of inorganic fertilizer application [13,212–214]. Overall, a reduction in soil NO_x emissions was observed in most of the experiments available in the literature but not in Reference [215]. It is worth noting that most of the experiments of biochar effects on soil NO_x emissions have been performed on vegetable cropping systems in China and requires more experimentations in other cropping systems and regions.

Table 3. Main controlling factors of the effect of biochar on soil nitrous oxide (N_2O) emissions.

Controlling Factors		Observations	Ref
Biochar	Feedstock	Lignocellulosic feedstocks (wood and crops) lead to significant reductions in soil N_2O emissions. Manures and other organic waste biochars vary in response.	[37,40,46]
	H/C_{org} ratio	Reduction in biochar H/C_{org} ratio increases mitigation of soil N_2O emissions. This is consistent with higher mitigation from lignocellulosic feedstock and higher production temperature having higher mitigation potential.	[37,39,46]
	Aging	Mitigation of soil N_2O emission by biochar is only transient, significantly decreasing after a year.	[37,38]
Soil	pH	Reference [46] found that mitigation of soil N_2O emissions by biochar is more pronounced under acidic and alkaline soil conditions, with the lowest mitigation potential under neutral soil pH. On the other hand, Reference [37] found that N_2O emission mitigation was lowest at soil pH of 6.5–7.0; Reference [40] found that there is little difference in soil N_2O mitigation across soil pH range but, for acidic soils (pH < 5), shows lowest potential and is nonsignificant.	[37,40,46]
	Texture	Mitigation of soil N_2O emissions by biochar increases from sandy texture toward finer textures, with maximum reduction in loams. However, clayey soils show the lowest mitigation potential. Soil texture responds differently under different soil moisture conditions.	[37,40,46]
	Moisture	Under high moisture, coarse soils show large variation in response to biochar with a mean negative mitigation potential of soil N_2O emissions, while other textures consistently reduce emissions. Under low moisture, fine soils show large variations in response to biochar, while other textures show consistently mitigation in soil N_2O emissions. After fertilization and under high soil moisture, biochar reduces soil N_2O emissions for about 1 month; after fertilization and under low soil moisture, biochar increases N_2O emissions for 3–4 days.	[37,46]
Management	Application rate	Increasing biochar application rate reduces N_2O emissions, with the maximum potential at about 90 t biochar/ha and above. Significant reductions are only observed at application rates above 10 t biochar/ha (∼1% application rate).	[37,40,46]
	Fertilizer	Biochar has more potential in decreasing soil N_2O emissions under fertilized conditions, particularly in fields. Biochar does not significantly reduce soil N_2O emissions from organic and ammonium nitrate fertilizer. However, it has a significant effect under urea and nitrate fertilization conditions.	[33,37,40]

H/C_{org}: Hydrogen to organic carbon in biochar (excluding carbonates in biochar's ash).

NH_3 volatilization increases with increase in biochar's alkalinity, in acidic soils and with low cation exchange capacity (CEC), and under large addition of biochar. Aging of biochar will mitigate increases in NH_3 emissions due to transient liming effect and increased biochar's CEC under aging (see Table S9 in SI based on References [46,47]).

Biochar increases plant available P in soils by 45% and microbial biomass P by 48% [48]. In a meta-analysis, it was found that biochar significantly increases phosphorus availability in soils for 5 years [216]. Biochars derived from manure and crop residues feedstock have higher content of P than other feedstock [49]. Biochar P is less mobile than agricultural residues P and could act as a slow-release P fertilizer. Biochar can be a P-recycling route from agricultural residues [50]. In terms of controls, crop residue and manure biochars increase soil P availability, less biochar-P is available at higher pyrolysis temperature, and alkaline soils may see P availability reduced (pH > 7.5) due to biochar's liming effect (see Table S10 in SI based on References [48,216]).

Leaching of nutrients (N and P) is affected by biochar as (1) it affect their availability in soils (see Tables S8, S10, and S11 in SI based on References [144,217]) and (2) it affects soil water regime in soils (see Table S12 in SI based on References [22–24,170,218–221]).

8.3. Water

Biochar improves soil water status by increasing the water holding capacity (more in coarser than finer soils) and by increasing the hydraulic conductivity in fine soil but by decreasing it in coarse soil [22,24]. Higher macroporosity and lower hydrophobicity of biochar increase soil water retention [170]. Interactions between biochar particles and soil aggregates are also important in modifying water holding capacity and water flow in soils by increasing or reducing soil interpore volume [22,219,220,222]. Better soil wet aggregate stability and soil consistency could also reduce pore clogging under wet conditions [22]. Biochar effects on soil water cycle also allow for increasing plant water availability [22,24,25]. Plant water use efficiency is also improved in certain studies but not always [25,92]. A minimum of 20–25 t biochar per hectare may be required to effectively increase soil available water capacity and to significantly modify soil hydraulic conductivity [22,24] (see Table S12).

8.4. Biophysical Effects

Biochar application will affect soil temperature via several interacting mechanisms: decreased soil albedo, increased soil moisture, reduction in soil volumetric heat capacity, conductivity, and diffusivity [22,223]. Patterns are different whether crop is present: biochar increases daily and seasonal soil temperature fluctuates in the absence of crops [224] and reduces it under cultivation [224–226]. Yan and colleagues [226] found that average soil temperature, average of the lowest and highest daily soil temperatures, and whole accumulated soil temperature is higher under biochar treatment, which may have consequences for soil carbon cycling, plant germination, and growth.

Biochar is a black material capable of absorbing light. Changes in light absorbance affect surface albedo. Genesio and colleagues [71] found that reflectance of soil biochar mixtures (30 and 60 t biochar/ha) decreased across all frequencies, while Zhang and colleagues [227] (4.5 and 9 t biochar/ha) found that reflectance in the short-wave domain (350–500 nm) was increased and decreased otherwise (500–2474 nm).

Changes in light absorbance affect surface albedo. Reduction in soil albedo due to biochar has been measured to be in range of 0.1 point [22]. At similar biochar application rate, decrease in soil albedo tend to be larger in lighter soils than darker ones [228]. However, soils with albedo of about 0.087–0.125, biochar's effect is not appreciable [228]. Soil albedo decreases with increasing biochar application rate [228], but tend to level off after a certain additional amount [71,225,228]. Crop canopy masks the effect of biochar on soil albedo [69,71] but not always completely [68,224]; the masking effect is related to the leaf area index [225]. Albedo of biochar amended soils is 32–58% lower under wet conditions compared to dry conditions [228]; biochar may also have an indirect effect on albedo as it increases soil moisture. Reduction in soil albedo can be mitigated by management choices: by tilling, which may mitigate the decrease in soil albedo after biochar application [71], though black spots on cropland with historical charcoal deposits (>120 years) are still visible [201]; by choosing crops with earlier canopy development [225] or cultivars with higher albedo [71]; or by using cover crops instead of leaving soils bare.

Biochar effects on soil water retention [22,24], soil water evaporation [229], and plant available water [22,24,25] also affect surface energy balance by affecting the partitioning of the incoming energy between sensible and latent heat [67]. Fischer and colleagues [25] also estimated that biochar increases evapotranspiration by about 5% in a coarse soil at about 150 t biochar/ha, as more water entering soil is stored and available to plants for evapotranspiration. Koide and colleagues [230] found an increase in 0.8–2.7 days of transpiration after biochar addition. Increased evapotranspiration would have a cooling effect.

9. Life-Cycle Assessment of Biochar Systems

We revised 34 studies performing life-cycle assessments (LCAs) of a biochar system with land application. Table 4 shows a summary of the most common assumptions and modeling approaches chosen by these studies, and Figure 3 shows a summary of the climate change impacts of biochar systems. Positive values in Figure 3 correspond to net emissions of GHGs, while negative values represent net avoided emissions of GHGs or sequestration of carbon (e.g., positive values indicates warming, while negative values indicate cooling). When possible, climate change impacts of biochar systems are shown across the main life-cycle stages, i.e., supply chain and pyrolysis, avoided emissions from coproducts, carbon sequestration in soils, and effects to soil emissions [69,123,151,152,231–235]. Some studies do not provide the required disaggregation of the data accross the different life-cycle stages, and in this case, only the total score is included in Figure 3 [236–248]. Some LCA studies define their functional unit as "per tonne of crop" or "per hectare cultivated" and did not provide conversion factors to transform their results into "per tonne of feedstock" as presented in Figure 3 [249–260]. As such, results from these studies are qualitatively discussed but not included in Figure 3. Studies are distinguished by feedstock type: herbaceous (like grasses, leafy or crop biomass), wood (forest residues or dedicated short-rotation coppice), and organic waste (manure or digestate) (Table S13).

Table 4. Main assumptions and modeling approaches for biochar system in life-cycle assessment studies.

Parameter	Typical Assumption	Ref
Biochar stability	15/85%, 20/80%, or 30/70% fraction of labile/recalcitrant fractions in biochar	[123,237,251,256]
	Remaining carbon in biochar after 100 year in soils: 68%	[69,151,152,231]
Reduced fertilizer use	Nitrogen: 7.2–10% and up to 25–30% reduction	[69,123,152,231–233]
	Phophorus: 5–7.2% reduction	[69,123,152,231,232,251]
	Potassium: 5–7.2% reduction	[69,123,152,231,232,251]
Reduced soil N_2O emissions	15 to 50% reduction in soil N_2O emissions; some studies model the transient effect of biochar on soil N_2O emissions; reduction of N_2O emission via reduced application of N fertilizer	[69,123,151,152,231–233,249]
Changes in soil CH_4 emissions/uptake	20% reduction in soil CH_4 emissions in paddy rice field; reduced upland soil methane sink by 0–50%	[151,251,252]
Effect on SOC	Changes in SOC through increase in NPP (5–10% increase) and negative priming on native SOC (5–10% decrease in decomposition rate); sensitivity analysis on SOC change from −12 to +21%	[152,231]
	Additional sequestration of 4 tC/ha over 30 years, 3.4 tC/ha over 25 years	[151,232]
Soil leaching	Reduced heavy metal leaching from soils	[244,245]
Functional unit	CO_2eq/kg feedstock	[69,123,152,231–233,244,245,248,256,258]
	CO_2eq/kg biochar	[237–240,254,259]
	CO_2eq/kg food produced	[249–252,257]
Biochar's yield effect	Modeled via the functional unit: increased yield lowers the yield-scaled GHG emissions intensity of food production	[251,257]
	Reduced fertilizer input for similar crop yield	[233,240]
	Increased NPP lead to more biomass output for biochar production or increases SOC	[152,231,241]
Pyrolysis coproduct treatment	Substitution; coproducts displace other products; associated burdens are substracted: electricity, residential, or industrial heat; various waste treatment options; cooking fuel	[69,123,125,152,231,232,236–238,241,252,257–259]
	Allocation, burden/benefits distributed across coproducts by mass, energy, or economic allocation	[239,254]
	Not treated; they are assumed to be outside system boundaries and to provide neither positive substitution effects nor burden	[239,240,242]

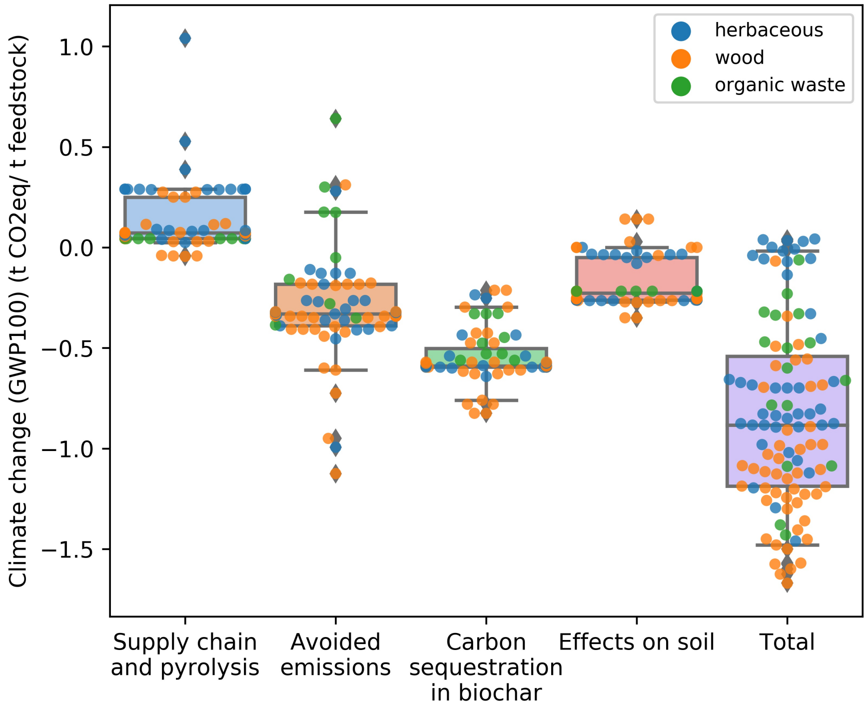

Figure 3. Survey of climate change impacts from life-cycle studies of pyrolysis systems with biochar production and application to agricultural fields: Positive values correspond to net emissions of GHGs, while negative values represent net avoided emissions of GHGs or sequestration of carbon (e.g., positive values indicates warming, while negative values indicate cooling). 'Supply chain and pyrolysis' refer to feedstock provision and pretreatment, pyrolysis, and transport; 'avoided emissions' accounts for avoided fossil carbon emissions by using bio-oil and pyrolysis gas for energy production and, in some cases, also accounts for avoided emissions due to reduced fertilizer consumption; and 'effects on soil' accounts for biochar effects on soil emissions (priming effect on SOC and NO_2 emissions) and changes in albedo. Each dot represents one biochar-production system, dots on the same line are results that uses the same assumptions. Boxes represent the 1st and 3rd quartile, with the middle line being the median; whiskers represent the 5th and 95th percentiles. This figure is based on References [69,123,151,152,231–248].

Supply chain and pyrolysis accounts for emissions occurring during feedstock collection, preprocessing (e.g., drying and chipping), and pyrolysis (e.g., start-up and exhaust gases). We found an overall climate impact of pyrolysis systems that ranges from net emissions of 1.04 to a net avoidance of emissions of −0.04 tCO_2eq/t feedstock (from 5th to 95th percentile). Negative values are due to studies that account for avoided emissions from degradation/burning of forest residues [231,233]. Use of dedicated crops leads to higher GHG emissions due to inputs required for their production (e.g., irrigation and pesticide use) (References [231,234,241] vs. Reference [152]) and, particularly, if indirect land-use change and loss of carbon are accounted for [123]. Use of crop residues can also lead to larger emissions of GHG from supply chains due to the allocation of part of the emissions from crop production to the residues [231]. Biomass conversion to biochar via pyrolysis usually has low emissions of GHG as syngas is used to run the conversion plant (avoiding fossil fuel input) and as biogenic carbon is usually assumed to be neutral. Transportation of feedstock and of biochar represents usually less than 10% of the GHG emissions from the supply chain [123,231–233,238], even under long transportation of feedstock [231,240] (e.g., from Canada to UK or from Indonesia to Australia). Fast pyrolysis with bio-oil upgrade to liquid fuel also leads to higher GHG emissions during the supply chain due to lower energy efficiency and more energy input required for feedstock processing [232]. Drying of wet feedstock (e.g., manures) can represent 25–83% of the supply chain's GHG emissions, particularly if heat production during pyrolysis or from coproducts is not enough to meet drying needs (requiring fossil energy) [237,246,247].

The use of forest or agricultural residues have implications for nutrient cycling and soil carbon, which are not always modeled in LCA. Wang and colleagues [232] included nutrient loss from feedstock collection, and Nguyen and colleagues [256] accounted for both nutrient and soil carbon loss, while Hammond and colleagues [231] accounted for avoided emissions during residue decomposition on forest ground. Changes in SOC stocks over consecutive years of residue removal can significantly increase the life-cycle emissions of the produced biofuels [153], and the same risks can occur for biochar production unless biochar is returned to the same field.

Avoided emissions account for saved emissions from avoided heat/electricity due to bioenergy production from pyrolysis coproducts and from fertilizer production due to higher fertilizer efficiency. Its contribution ranges from GHG emission savings of 1.13 tCO_2eq/t feedstock to a net emission of 0.64 tCO_2eq/t feedstock. GHG intensity of the background energy system being substituted is an important factor for controlling the size of avoided emissions. For example, Azzi and colleagues [151] found that, at a GHG intensity of the electric grid of 1 $kgCO_2eq$/kWh, biochar's coproducts can offset by ∼0.25–1 tCO_2eq/t feedstock while, at 0.2 $kgCO_2eq$/kWh, the offsets are only ∼0.1–0.25 tCO_2eq/t feedstock. The positive values under 'avoided emissions' in Figure 3 occur when pyrolysis replaces waste treatments that save large amounts of GHG emissions (for example, recycling of cardboard), so that diverting the waste stream results in less avoided emissions overall [152]. Wood contains more energy than herbaceous feedstock, leading to higher fossil fuel savings due to higher bioenergy production (∼25–38% increase from crop to wood residues) [231]. Large-scale plants have lower energy losses, leading to higher energy output and fossil fuel savings (∼122–150% increase from plant treating 2000 to treating 100,000 tonnes of feedstock per year) [231]. Increasing pyrolysis temperature decreases biochar yield and increases bio-oil and syngas yields; 3.75 more energy is recoverable at 600 compared to 300 °C [247].

Some LCA studies compare biochar systems to alternative uses of the same feedstock. For example, Clare and colleagues [236] showed that using straw residues for gasification or coal briquettes leads to higher climate change mitigation potentials than biochar in China due to larger offsets of fossil energy. Other studies [69,151,253] reach similar conclusion with standard bioenergy systems achieving more climate benefit compared to biochar sequestration. As a general interpretation, larger climate benefits from using residues for biochar production are achieved in regions with low carbon intensity energy systems, whereas in regions with high carbon intensive energy systems, the use of residues for bioenergy is a better option as it can bring the largest emission savings.

Carbon sequestration accounts for biochar sequestration in soils, and the variation in the results is mostly due to different assumptions of biochar stability. As seen in Table 4, some studies assume a carbon labile fraction of 30% [251,254], 20% [123,236–238], or 15% [69,152,231,232]. Some studies then assume a mean residence time (MRT; between 200–500 years [69,231–233]) of biochar and integrate the loss over 100 years [69,231–233]. Other studies assume that only the labile fraction is degraded to CO_2 [236,251]. In Figure 3, higher biochar stability means more carbon sequestration. However, there is a large difference between the assumption of stable carbon in LCA studies and the 3%/97% split noted by Wang and colleagues, which is based on a meta-analysis of field/laboratory experiments [20]. This may be due to a lack of compiled carbon stability data at the time of the LCA studies as the meta-analysis of biochar stability are from 2016 and 2018 [18,20] or to a choice of keeping conservative assumptions. Thers and colleagues [249] showed that carbon sequestration is increased at higher pyrolysis temperature by 10% from 400 °C to 800 °C.

Rapid decomposition of the labile fraction of biochar and burning of bio-oil/syngas leads to carbon emissions that can happen before biomass had time to regrow. This delay between emission of biogenic carbon and biomass regrowth leads to a temporary increase in atmospheric carbon concentration that has a warming effect [261,262]. This temporary effect is found to reduce the climate benefits of biochar systems of small round wood (regrowth cycle 50 years) and crop residues (regrowth over one year) by about 15% [69].

Effects on soils account for biochar's effects on soil emissions, soil albedo, and native SOC. Biochar effects on soil emissions are usually modeled crudely and are likely to depend on local soil conditions. Reduction in soil N_2O emissions account for about 1.5–4% [123,232,233,252] of the total climate mitigation of biochar. Changes in soil CH_4 emissions/uptake have a warming effect of −1.1% to a cooling effect of +9% of the overall climate impacts [151,252]. Azzi and colleagues [151] found that biochar can reduce GHG emissions up to 14–23% of its total mitigation contribution. They modeled a cascading effect of biochar, where it is used as cattle feed addition (lowering enteric methane emissions) and is added to manures during storage (lowering N_2O, CH_4, and NH_3 emissions) before its land application (lowering N_2O emissions). Reduced soil albedo decreases climate mitigation benefits by 13–22% [69].

Aspects like changes in soil albedo, evapotraspiration, and NTCFs emissions are difficult to quantify, and an estimation of the climate effects usually requires coupled land–atmosphere climate models to account for complex interactions between precipitations and latent heat, surface radiation, and clouds [263,264]. NTCFs have very short lifetime in the atmosphere, leading to spatially heterogeneous concentrations, and affect climate forcing through multiple pathways, making their effects on climate uncertain [64]. As such, those effects are not streamlined in the LCA methodology but can be included in some specific case studies. For example, References [265,266] showed that albedo effect can be significant, offseting partially or even completely the lifecycle GHG emissions of biofuel production. Arvesen and colleagues [267] found that albedo changes and that cooling aerosols offset 60–70% of life-cycle GHG emissions of boreal foerest bioenergy. In addition, changes in evapotranspiration from conversion from annual to perenial cropping systems can offset 0.5 °C of warming, according to Georgescu and colleagues [266].

The various assumptions and modeling approaches lead to a large variation in the overall climate change mitigation potential of biochar (see 'Total' in Figure 3). Some studies found an overall warming potential of biochar despite the amount of carbon added to soils. These studies [239,240,242] do not consider carbon abatement from pyrolysis coproducts and do not consider low biochar yield and stability. Looking at the total, appreciation of the carbon abatement differences between the feedstocks is easier, with usually the lowest climate change mitigation potential for organic wastes, followed by herbaceous and higher climate change mitigation potential for woody materials. The largest climate change mitigation potentials are found for woody feedstock and low pyrolysis temperature (∼300 °C) [238,247]. However, these two studies suffer from incomplete modeling of the effect of pyrolysis temperature on biochar stability. Their results must be contrasted by results from Thers and colleagues [249], who found a higher climate mitigation potential for high temperature biochar in their more thorough modeling of the effect of pyrolysis temperature (decrease in biochar yield but increase in biochar's carbon content and its stability and increase in bio-oil yield).

In terms of other LCA impact categories, some LCA studies focus on mid-point indicators and other go to end-points (Table S14). Pyrolysis systems for energy and biochar application to soils are found to have lower negative effects on human toxicity and eutrophication categories due to lower fertilizer and natural gas use [238,256]. Management of dedicated feedstock plantation for biochar production can increase risks of acidification and eutrophication and, therefore, on ecosystem quality due to use of fossil fuel in machinery, fertilizers, and pesticides [237,241]. Biochar produced from miscanthus plantation is found to provide only benefits in terms of climate change [260]. Handling of sewage sludge with biochar production and land application reduces the risks associated with their incineration and further landfilling or land application of their ashes, such as reduced aquatic ecotoxicity, carcinogenic and noncarcinogenic toxicity, acidification, and terrestrial ecotoxicity [244,245]. Biochar liming effect, lower fertilization need, and immobilization of heavy-metals in biochar are the mechanisms behind the reduced environmental load. In terms of end-points, energy requirements for drying is an important contributor of degradation of ecosystem ecosystem quality and human health [237]. Traditional kilns for biochar production can also present a risk for human toxicity via increased particulate matter exposition [257–259].

However, not all effects of biochar on soil are modeled, as discussed in Section 4. Nitrogen emissions are important for ecosystem quality and human health, but effects of biochar on soil NO_x and NH_3 are not modeled. Leaching of nutrients are reduced but only via the indirect effect of the reduced need for fertilizer and not because of higher retention due to biochar addition.

Some LCA studies take into account the positive effect of biochar on yields. Choosing the functional unit as unit of food or unit of land, they implicitly model biochar's yield effect, as environmental impacts become divided over larger output [251,257]. Peters and colleagues modeled climate change mitigation potential of short-rotation coppice for biochar production using a functional unit per unit of area [241]. Biochar is reapplied to the plantation, increasing yield and biomass output and further improving the plantation climate mitigation potential. Two studies also model biochar's effect on NPP by assuming that it would increase accumulation of plant carbon in soils [152,231].

10. Biochar and Social and Ethical Aspects

Justice is at the heart of mitigating climate change, whether it is to protect future generations from unstable, extreme climate or to protect the most vulnerable that have less responsibility in global warming but may suffer the most consequences. Technologies aiming at limiting warming are also subject to a set of ethical considerations, among them fairness and justice [268]. An aspect that is particularly important is the distribution of potential burden of deploying certain technologies, especially toward the most vulnerable populations.

Negative emission technologies are subject to multiple ethical considerations. Among NETs, biochar is one with the lowest ethical side effects. Low input agriculture on small-scale farms is widespread in tropical and subtropical regions [15]. Yield response to biochar is more pronounced in those regions with weathered soils [29]. Biochar may provide important social benefits for some of the most vulnerable farmers. Higher soil water retention may also provide adaptation to climate change in some of the most vulnerable regions. Higher yield and retention of soil fertility may also help mitigate shifting agriculture practices in tropical forests, which is responsible for about 24% of forest disturbances [269].

In terms of its deployment being a local practice, it is possible to develop strategies and protocols where it can be applied only when negative side effects are reduced. There is a risk that a large-scale deployment of biochar technologies, such as at the scales required by several climate change mitigation scenarios, can have adverse side effects on food security or natural ecosystems due to expansion of dedicated biomass plantations for biochar feedstocks at expenses of croplands or forests. Cross-sectoral integrated policies should ensure that future growth of a biochar market would not lead to competition with food production or trigger deforestation. Further, biochar is intended to remain decades or even centuries in soils, and its long-term aging and effects on soils are not fully understood. Removing biochar once in soils seems hypothetical and would require important soil disturbance.

11. Conclusions

In this review study, we discussed implications of biochar application to soils around three areas of concern: climate, food security, and ecosystem and toxicity. We provided some key controlling factors regarding biochar effects on those areas of concern. However, most of the identified patterns on biochar's effects on soil are gathered from meta-analyses, which sometimes may suffer from methodological issues, such as combined nonindependent data points, raising question of pseudo-replications that may have overestimated or increased the confidence of some effects of biochar in soils [36,38,270,271].

Biochar is an attractive NET for CDR as it can supply two marketable products, biochar as soil amendment and bioenergy generation via biochar coproducts (bio-oil and syngas). Pyrolysis is a rather simple, known technology that can be deployed in both developed economies and developing countries. More agricultural benefits associated with biochar systems are expected in developing countries with low agricultural inputs and degraded/weathered soils. Low investment potential can limit the additional climate mitigation benefits from bioenergy production as it requires more infrastructure to recover, produce, and distribute energy. On the other hand, developed economies in temperate regions may expect less agricultural benefits from biochar application to soils, but higher level of investment can allow to avoid fossil energy use and to provide incentives for negative emissions.

Climate change mitigation benefits of biochar are potentially large but depend on soil interactions, its production conditions, availability of cheap and sustainable feedstocks, and management practices. As another biomass-based NET, biochar supply is constrained by the availability of forest or crop residues or of land to grow dedicated bioenergy crops. Interactions of biochar with the climate systems are more complex than carbon sequestration only or reductions in GHG emissions from soils. They include changes in surface albedo, soil water fluxes, and emissions of NTCFs, which are difficult to quantify and affect the estimates of net local and global climate effects of biochar systems.

Some aspects of biochar needs further investigation. Potential emissions of black carbon from soil after biochar application would have implications for climate mitigation and toxicity, and the availability of specific emission factors would facilitate their inclusion in environmental assessment studies. Leaching of dissolved biochar carbon and its degradation in water systems could decrease its long-term sequestration potential while being toxic. Interactions between the biophysical and biogeochemical effects of biochar in soils are highly complex. Future integration of biochar deployment scenarios with climate models of varying complexity can offer an opportunity to quantify biochar's climate interactions in full and can distill simplified metrics to be used in individual studies aiming at assessing the role of biochar for climate change mitigation and adaptation in different geographical contexts and for different feedstock-application combinations.

Supplementary Materials: The following are available online at http://www.mdpi.com/2073-445X/8/12/179/s1, Table S1: Main controlling factors on the effect of biochar on agricultural yield; Table S2: Main controlling factors on the effect of biochar on soil and exposition to toxic compounds; Table S3: CDR requirements for different temperature pathways; Table S4: Total cumulative CDR requirements for different temperature pathways; Table S5: Key biochar properties and their controlling factors under slow pyrolysis; Table S6: Main controlling factors of the effect of biochar on SOC priming; Table S7: Main controlling factors of the effect of biochar on soil methane (CH_4) emissions or uptake; Table S8: Main controlling factors on the effect of biochar on soil nitrogen availability; Table S9: Main controlling factors of the effect of biochar on soil ammonia (NH_3) volatilization rate; Table S10: Main controlling factors on the effect of biochar on soil phosphorus availability; Table S11: Main controlling factors on the effect of biochar on soil nutrient leaching; Table S12: Main controlling factors on the effect of biochar on soil water availability and soil hydraulic conductivity; Table S13: Classification of the LCA studies under type of feedstock and origin (residue, dedicated plantations, or waste) of which the life-cycle stage results were used in Figure 3 in the main text; Table S14: Classification of LCA studies according to the type of impact/indicator they include in the analysis.

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Abbreviations

The following abbreviations are used in this manuscript:

BECCS	Bioenergy with Carbon Capture and Storage
C/N	Carbon-to-Nitrogen ratio
CDR	Carbon Dioxide Removal
CEC	Cation Exchange Capacity
DACCS	Direct Air Carbon Capture and Storage
DOC	Dissolved Organic Carbon
GHG	Greenhouse Gas
H/Corg	Hydrogen to Organic Carbon ratio (excludes carbonates in ash)
K	Potassium
LCA	Life-Cycle Assessment
N	Nitrogen
NET	Negative Emission Technology
NMVOC	Non-Methane Volatile Organic Carbon
NPP	Net Primary Productivity
NTCF	Near-Term Climate Forcer
P	Phosphorus
PAH	Poly-Aromatic Hydrocarbon
PM	Particulate Matter
SI	Supplementary Information
SIC	Soil Inorganic Carbon
SOC	Soil Organic Carbon
VOC	Volatile Organic Carbon

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Chapter 3. Life-cycle assessment to unravel co-benefits and trade-offs of large-scale biochar deployment in Norwegian agriculture

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Life-cycle assessment to unravel co-benefits and trade-offs of large-scale biochar deployment in Norwegian agriculture

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ABSTRACT

Limiting temperature rise below 2 °C requires large deployment of Negative Emission Technologies (NET) to capture and store atmospheric CO_2. Compared to other types of NETs, biochar has emerged as a mature option to store carbon in soils while providing several co-benefits and limited trade-offs. Existing life-cycle assessment studies of biochar systems mostly focus on climate impacts from greenhouse gasses (GHGs), while other forcing agents, effects on soil emissions, other impact categories, and the implications of a large-scale national deployment are rarely jointly considered. Here, we consider all these aspects and quantify the environmental impacts of application to agricultural soils of biochar from forest residues available in Norway considering different scenarios (including mixing of biochar with synthetic fertilizers and bio-oil sequestration for long-term storage). All the biochar scenarios deliver negative emissions under a life-cycle perspective, ranging from -1.72 ± 0.45 tonnes CO_2-eq. ha^{-1} yr^{-1} to -7.18 ± 0.67 tonnes CO_2-eq. ha^{-1} yr^{-1} (when bio-oil is sequestered). Estimated negative emissions are robust to multiple climate metrics and a large range of uncertainties tested with a Monte-Carlo analysis. Co-benefits exist with crop yields, stratospheric ozone depletion and marine eutrophication, but potential trade-offs occur with tropospheric ozone formation, fine particulate formation, terrestrial acidification and ecotoxicity. At a national level, biochar has the potential to offset between 13% and 40% of the GHG emissions from the Norwegian agricultural sector. Overall, our study shows the importance of integrating emissions from the supply chain with those from agricultural soils to estimate mitigation potentials of biochar in specific regional contexts.

1. Introduction

The achievement of the Paris agreement of limiting global temperature rise to well below 2 °C is likely to require large amount of carbon dioxide removal (CDR) (Rogelj et al., 2018). Depending on temperature pathways, 95% of the estimated cumulative need for CDR falls between 130 and 1600 GtCO2 (Huppmann et al., 2018; Rogelj et al., 2018). Several options have been proposed as negative emission technologies (NET) for CDR: afforestation and reforestation, soil carbon sequestration, biochar, bioenergy with carbon capture and storage (BECSS), direct air capture, enhanced weathering and ocean fertilization, among others (Minx et al., 2018).

Biochar is produced from thermo-chemical conversion of biomass in absence of oxygen and it is considered a NET because it is a stable carbon-based product that can be stored in soils for centuries (Smith, 2016). Depending on the future socioeconomic scenarios and temperature targets considered, biochar can provide from 10 to 35% of the required CDR deployment rate in 2050 (Tisserant and Cherubini, 2019). Biochar production can rely on today's non-used resources, like forest and crop residues, and it has several co-benefits. For example, it produces useful co-products, such as non-condensable gasses and bio-oil (a mixture of organic compounds and water) (Crombie and Mašek, 2015; Woolf et al., 2014). The technology is well known and easy to implement, although large facilities are still lacking (Minx et al., 2018). Bio-oil, which is also rich in biogenic carbon, could be stored in geological deposits to further improve the CDR potential of biochar (Schmidt et al., 2018; Werner et al., 2018). There is also evidence of a series of positive effects of biochar use in agriculture, such as increases in plant yields (Jeffery et al., 2017), reduction of N_2O emissions and nitrogen leaching from soils (Borchard et al., 2019; Liu et al., 2019),

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improved soil water retention (Razzaghi et al., 2020), restored soil fertility, prevention of land degradation (Ali et al., 2017; Saifullah et al., 2018; Yu et al., 2019), and remediation of contaminated sites (Abbas et al., 2018; Yuan et al., 2019; Zama et al., 2018). Biochar is thus attracting increasing attention as one of the most promising options to achieve large-scale CDR deployment and simultaneously co-deliver improvements on multiple sustainability issues (Semida et al., 2019; Smith et al., 2020; Tisserant and Cherubini, 2019).

Assessing the climate change mitigation potential and the environmental sustainability profile of a technology requires a life-cycle perspective that accounts for direct and indirect emissions along its value chain. Life-cycle assessment (LCA) is a useful method to monitor potential co-benefits or trade-offs by tracking several environmental impacts. Many LCA studies of biochar application to agricultural soils have been performed over the years and have been reviewed in two recent literature reviews (Matuštík et al., 2020; Tisserant and Cherubini, 2019). All studies generally converge on the net climate mitigation benefits of biochar, but the magnitude depends on a variety of factors such as type of biomass feedstocks, pyrolysis conditions, biochar treatment, agriculture management and methodological assumptions. Results are thus highly case-specific. Most of the existing studies mainly assessed the climate effects using the Global Warming Potential (GWP) with a time horizon (TH) of 100 years as the default characterization factor (or emission metric), and only consider impacts from greenhouse gasses (GHGs), mainly CO_2, CH_4 and N_2O. This approach has limitations because on the one hand it ignores multiple temporal dimensions of the climate system response to emissions (e.g., either in the short-term or in the long-term), and on the other hand it does not take into account the climate change effects of the so-called near-term climate forcers (NTCFs), such as aerosols (SO_x, black carbon (BC), organic carbon (OC)) and ozone precursors (NO_x, non-methane volatile organic compounds (NMVOC), CO), which cause a strong but time-limited perturbation to the climate (Cherubini et al., 2016; Jolliet et al., 2018; Levasseur et al., 2016a). Further, recent literature reviews noted that analysis of other impact categories besides climate change is limited, and argued future studies should include an assessment of effects in other environmental areas of concerns that are relevant for biochar production and use (Matuštík et al., 2020; Tisserant and Cherubini, 2019). For example, despite its clear importance, only a few LCA studies include biochar's effects on soil emissions (Azzi et al., 2019; Field et al., 2013; Roberts et al., 2010; Thers et al., 2019; Wang et al., 2014). Biochar can potentially affect nitrogen emissions from soils like N_2O, ammonia volatilization, NO_x, and nitrogen leaching (Borchard et al., 2019; Liu et al., 2019; Pourhashem et al., 2017), but the influence of these biochar-induced changes for a range of environmental impact categories has not yet been explored within a life-cycle perspective. These emissions, together with other NTCFs, are important drivers of air quality, eutrophication, or acidification. Similarly, only some LCA studies include positive effects of biochar on yields and nutrients, by either modeling increase in food production or reduction of fertilizer inputs (Field et al., 2013; Mohammadi et al., 2016; Robb and Dargusch, 2018; Sparrevik et al., 2013).

In Norway, increasing soil carbon stock is an important strategy from a climate perspective and for soil health and food production, and biochar has been identified as one of the technologies with the highest potential (Rasse et al., 2019). Norway has large amounts of forest residues that are left unused after extraction of commercial roundwood or from wood industries (Cavalett and Cherubini, 2018), and they are a promising feedstock for biochar production to stimulate a circular economy perspective and reduce pressure on terrestrial ecosystems. In this study, we assess the life-cycle environmental sustainability effects of alternative scenarios of large-scale deployment of biochar production from forest residues and application to agricultural soils in Norway. Biochar production is modelled using a process simulation software to derive emission factors and the mass and energy balance. Different biochar scenarios are investigated, and they differ by the type of biochar used as soil amendment in agriculture (untreated biochar or a biochar-fertilizer mix), and use of biochar co-products (production of heat and power or pumping bio-oil into geological storages to maximize carbon sequestration). The analysis focuses on grain production (barley) and quantifies the environmental impacts from both the life-cycle stages and the changes in soil emissions under Norwegian conditions of biochar use in agriculture. Co-benefits and trade-offs are explored for a range of impact categories: climate change, stratospheric ozone depletion, fine particulate matter formation, tropospheric ozone formation, terrestrial acidification, marine eutrophication and terrestrial ecotoxicity. Multiple climate metrics are used to assess climate change mitigation benefits across different time dimensions, and effects of both GHGs and NTCFs are considered. The overall robustness of the results is evaluated with a Monte-Carlo analysis (10 000 simulations) that considers a variety of uncertainty ranges in key process parameters, modeling assumptions, emission factors, and climate metrics (especially NTCFs). The climate change mitigation potential and other environmental sustainability effects of large-scale biochar deployment in Norway are quantified both per individual process unit (e.g., hectare of land, kg of biochar, or kg of grain) and for a national large-scale deployment (i.e., per year), so to estimate the overall mitigation potentials and side-effects.

2. Methods

The methods section is structured as follows: Section 2.1 presents the system boundaries and an overview of the reference system and the different scenarios; Section 2.2 describes the reference system; sections from 2.3 to 2.6 introduce the modeling of the various aspects of the biochar scenarios (i.e. feedstock collection and transport, pyrolysis, biochar-fertilizer production and application to soil); Section 2.7 presents the effects of biochar on soil; Section 2.8 explains the different climate metrics and impact categories considered for the analysis; Section 2.9 presents the approach to scale up the analysis of the potentials and effects of large-scale biochar application in Norway; Section 2.10 describes the uncertainty analysis.

2.1. System boundaries and biochar scenarios

Fig. 1 shows an overview of the scenarios and system boundaries for the life-cycle assessment of biochar production and application to agricultural soils in Norway. The analysis compares grain production in Norway without or with biochar application.

The reference system includes farming activities (ploughing, fertilization, pesticide application) and inputs (fertilizers, machineries, lime) required for the management of one hectare of land producing barley over the period of one year without addition of biochar to soil.

The reference system is compared to four scenarios where biochar produced from forest residues is spread on land, while the other farming activities remain the same (unless those affected by biochar, such as changes in fertilizer management and soil emissions). The four biochar scenarios are: (i) "biochar", where biochar is directly applied to agricultural soils and biochar co-products are burnt to provide heat for pyrolysis and feedstock drying (no use of the extra heat available); (ii) "biochar-fertilizer", where biochar is grinded and mixed with inorganic fertilizers and pelletized before its application to soils, and biochar co-products are burnt to provide heat for pyrolysis and feedstock drying (no use of the extra heat available); (iii) "biochar-fertilizer with CHP", as in (ii) but co-products are burnt in a CHP unit to meet the electricity and heat demand of the pyrolysis plant, and the excess energy is assumed to displace electricity from the grid and heat from natural gas; (iv) "biochar-fertilizer with bio-oil sequestration", where biochar is treated as in (ii) and all the syngas and part of the bio-oil are combusted to provide heat for pyrolysis, and the remaining of the bio-oil is recovered, transported and pumped into off-shore geological deposits to maximize carbon storage.

Biochar is assumed to be produced by three large-scale facilities

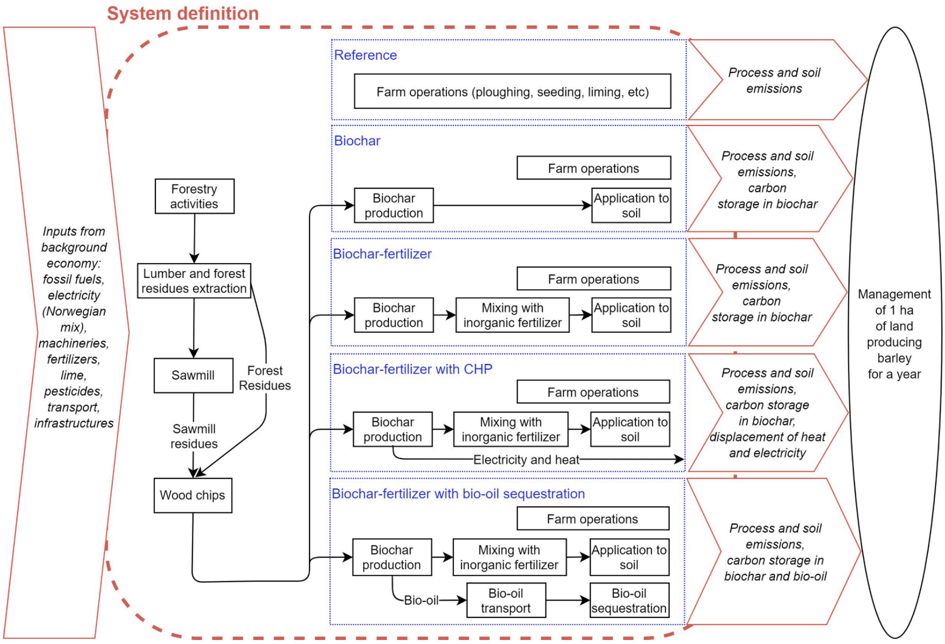

Fig. 1. Overview of the system boundaries and biochar scenarios.

located in Oslo, Stavanger and Trondheim. Biochar supply chain starts with the provision of the feedstock to the plants and includes forestry activities and extraction of forest residues. Residues from the wood industry are also included as potential feedstock. Biochar's effects on soil include changes in N_2O, NH_3 and NO_x emissions, changes in nitrogen leaching, and in the case of biochar-fertilizer application, a positive effect on yield is considered. If not indicated otherwise, Ecoinvent 3.5 (Wernet et al., 2016) was used to gather emission inventories, energy consumption and emission factors associated with the provision of equipment, materials and inputs.

2.2. Reference system

The reference system is the management of one hectare (one complete crop cycle) for one year producing barley, which is the main grain produced in Norway on about 50% of the total grain area (SSB, 2020a). We used reported yields data of barley in Norway from the official national statistics (SSB, 2020b), and estimated an average barley yield of 3756 kg ha^{-1} over the 2009–2018 timespan, with a standard deviation of 495 kg ha^{-1} (here assumed as a proxy of variability in terms of climate and location). Barley production is modeled by adapting the ecoinvent process for barley production in Germany (given on kg barley basis) to Norwegian practices. Field work follows common practices on Norwegian farms and includes ploughing, sowing, harrowing and leveling with stone picking, fertilizing, rolling, pesticide application (typically two applications per year, plus a chemical fallow every three years) and liming (250 kg CaO equivalent per year) (Henriksen and Korsæth, 2013). Fertilizer requirements per year are based on Norwegian average inorganic fertilizer application for barley: 127.5 kg N ha^{-1}, 17.3 kg P ha^{-1}and 63 kg K ha^{-1} (Gundersen and Heldal, 2013; Kolle and Oguz-Alper, 2018). Pesticides application follows typical Norwegian practices for barley (Aarstad and Bjørlo, 2019) and the fields are not irrigated. The inventory is available in Table S1.

2.3. Biomass collection and transport

Feedstock availability and life-cycle inventory for collection, processing and transport follows the model developed in a previous work (Cavalett and Cherubini, 2018). The model is based on county and species-specific production of commercial roundwood removals in Norway over the period 2011–2016. The amount of residues extractable is calculated using age-dependent and species-specific biomass expansion factors to quantify the amount of biomass left in forest after harvest (Lundmark et al., 2014). It is common practice in Norway to leave all forest residues in the forest due to a lack of market for utilizing branches and low-quality wood. In the country, forest residues typically represent a promising feedstock to enhance renewable material supply at no additional pressures from expansion of harvest and to revitalize rural areas through increased circular economy. A residue extraction rate of about 34% is assumed in our analysis, based on sustainable rates of extraction in other Scandinavian countries, where the utilization of forest residues is more common than in Norway (de Jong et al., 2017; Lundmark et al., 2014). A potential of 1.14 Mtonnes year^{-1} of forest residues is estimated, to which we can add an additional 0.56 Mtonnes year^{-1} of by products from the wood industry. Overall, about 82% of forest wood residues are from spruce, 17% from pine and 1% from birch. Life-cycle inventories for feedstock supply include the complete biomass value chain and account for inputs and emissions from harvesting, transport, chipping and processing of forest residues and wood industry residues in Norway. Norwegian-specific data for forestry operations and logistics were used (Cavalett and Cherubini, 2018).

Feedstock transport to the biochar conversion plants is modeled by assigning residues in each county to the nearest biochar conversion plant, after satisfying an equal share of forest residues to the three conversion plants. It is also assumed that the lumber output from forestry is treated within the same county, and the same transport distance is assumed for wood industry residues to the conversion plant. The distance from the county's capital to the conversion plant is used to estimate truck transport distances, or it is assumed to be 40 km if residues are located within the same county of the plant. Distances are weighted by the county's share of feedstock produced and a weighted average transport distance of 190 km from forest to plant is estimated at national level. It is assumed that the feedstock is transported at 40% moisture.

2.4. Pyrolysis

Inventories for biochar production are estimated by modeling the pyrolysis process in Aspen Plus process simulation software. The approach chosen is to model the feedstock biomass, biochar and tar (i.e. organic fraction of the bio-oil, which is a mixture of organic compounds and water) as non-conventional components, while syngas is modeled as a mixture of gas species. For modeling the pyrolysis reaction, a simple approach of converting the feedstock into products using yields is used. The pyrolysis is modeled at 500 °C, and the mass (carbon) yields are 28% (45.7%) to biochar, 56% (42.6%) to bio-oil, and 16% (11.7%) to syngas.

Non-conventional components modeling in Aspen plus requires the proximate analysis (i.e., composition in moisture content, fixed matter, volatile matter and ash content), the ultimate analysis (i.e., content in C, H, O, N, S, Cl) and the sulfate analysis (i.e., content in different forms of sulfur pyritic, sulfate and organic). These data are shown in Table S2 in the supplementary information (SI). The feedstock is modeled as spruce wood, whose composition is taken from the Phyllis2 database (phyllis.nl). Elemental composition and lignin content are taken from the average of the 43 samples in the database for Spruce. Fixed matter, volatile matter and ash contents are also taken from the same database. Biochar yield is determined as function of pyrolysis temperature and feedstock lignin content, and the yield of CH_4, CO, H_2 and C_2H_2 are estimated from regressions based on pyrolysis temperature (Woolf et al., 2014). Tar, CO_2 and water yields are determined from elemental mass balance. N can volatilize as HCN and NH_3 during pyrolysis, S as H_2S and Cl as HCl, CH_3Cl or KCl. Figures S1-S3 in the SI show regression analysis based on literature data of the share of conversion rates of N, S, Cl from the feedstock into different gasses as a function of temperature. These regressions are used to estimate the yield of these gas species for the specific temperature of our pyrolysis system.

For the ultimate analysis of biochar and tar, C, H, and O compositions are estimated from pyrolysis temperature and C, H, O content of the feedstock, using regressions from (Woolf et al., 2014). N content of biochar is assumed to be 0.1% (Morales et al., 2015). S and Cl content in biochar are determined from regressions in Figures S1-S3 in the SI. Tar is used to balance N, S and Cl elements. For the proximate analysis, it is assumed that all feedstock ashes remain in the biochar, which has a fixed matter content of 80% (Weber and Quicker, 2018) and volatile matter is determined to complete the balance. The proximate analysis of the tar (supposed ash-free) is determined using the average value for fixed and volatile matter for bio-oils (given on a dry basis) in the Phyllis2 database: 33.2% for fixed matter, 66.8% for the volatile matter.

The composition of the biomass, biochar and tar and the yields of the different products of pyrolysis are shown in the Tables S2 and S3 in the SI. Description of the Aspen Plus simulations is available in the supplementary text 1 together with Aspen Plus flow charts (Figure S4 and Figure S5) in the SI.

In the case of pyrolysis with combined heat and power (CHP) production, the tar and syngas are burned for recovery of electricity and heat at 28.5% and 71.5% of efficiency, respectively, in line with standard values for steam cycle CHP (Sipilä, 2016).

In the case of biochar production with bio-oil recovery for geological storage, part of the tar (11%) is used for combustion with syngas to produce the required heat for the pyrolysis plant to avoid relying on external fossil fuel. The rest of the bio-oil is transported to Stavanger and transferred to a tanker for transport of 400 nautical miles (one-way) (Gassco, 2017). Infrastructures required for pumping the oil to geological deposit is estimated from Ecoinvent process of offshore petroleum and gas production.

Electricity consumption for drying the feedstock and the pyrolysis reactor are taken from a model of biomass torrefaction (Manouchehrinejad and Mani, 2019), and energy requirements for blowing air for the combustion are given by Aspen Plus. Drying of wood is associated with emissions of NMVOC, estimated at 56 mg/kg biochar produced (Granström, 2009). In the case of the CHP, the energy requirement for producing the biochar-fertilizer is taken by the electricity output from the CHP, and it is thus subtracted from it. Similarly, the heat required for drying the feedstock is subtracted from the heat from the CHP. For the other cases, electricity consumption for producing the biochar or biochar-fertilizer is assumed to be from the Norwegian electricity mix from ecoinvent database (Wernet et al., 2016).

Aspen Plus-derived emissions from the pyrolysis-CHP system are complemented with emission factors measured from a medium scale pyrolyser (Sørmo et al., 2020). They include emission factors for polycyclic aromatic hydrocarbon (PAHs), NMVOC, PM10 and heavy metals associated with particulate matter (As, Cd, Cr, Cu, Pb, Hg, Mo, Ni, Sn). In the case of the pyrolysis with bio-oil recovery, the emission factors are corrected by the amount of tar sent to combustion.

The inventories for the different biochar production scenarios are shown in Table S4, and for the sequestration of bio-oil in Table S5.

2.5. Biochar-fertilizer

In the biochar scenario, biochar is directly applied to the field as a biochar soil amendment. In the biochar-fertilizer scenario, biochar is mixed with fertilizers before application to the soils to form the so-called biochar-based fertilizer (BCF). BCF is produced by grinding biochar into fine particles, then mixing them with a fertilizer and then pelletizing into a final product. Applying biochar in the form of BCFs is found to improve effects on yield and nitrogen use efficiency (Chew et al., 2020; Liu et al., 2020; Shi et al., 2020). Such an expected effect is especially important in Nordic conditions where biochar alone does not necessarily increase yields (O'Toole et al., 2018). Biochar has been shown to substantially reduce N_2O emissions, but this effect is more pronounced the first year after application (Borchard et al., 2019). For this reason, annual applications of biochar mixed with nitrogen fertilizer is expected to maximize the reduction in N_2O emissions (Guenet et al., 2021). Positive interactions between the carbon structure of biochar and nitrogen fertilizer in BCF are also expected to reduce NO_3^- leaching and thereby increase nitrogen use efficiency (Guenet et al., 2021). These positive effects of BCFs on nitrogen use efficiency and yield result from the slow release to the soil of the nitrogen absorbed on the biochar structure (Ibrahim et al., 2020). However, there are physico-chemical limits to how much nitrogen can be absorbed on a biochar structure. Most studies report nitrogen-sorption for biochar below 20 g N per kg biochar (Zhang et al., 2020), but we hypothesized that above-average products would be developed and selected towards a realistic upper value of 50 g nitrogen per kg biochar, which is still lower than several high values reported in the literature (Zhang et al., 2020). Our working hypothesis translates into 50 kg nitrogen per tonne of biochar, which implies that 2552 kg of biochar per hectare need to be applied as BCF to fulfill the nitrogen fertilizing requirements of a barley cropland in Norway. As softwood biochar has 0.51% K_2O available to plants (Ippolito et al., 2015), this reduces the need for potassium by 10.7 kg. The final loading of fertilizers to biochar to fulfill barley's requirements is thus 50 kg N, 6.75 kg P and 20.5 kg K per tonne of biochar.

Energy requirements for grinding and pelletizing the biochar is taken from (Manouchehrinejad and Mani, 2019). Due to lack of data for grinding the fertilizers, the same energy requirement of biochar per unit of (dry) mass is assumed. The total energy requirement is 0.21 kWh per kg biochar-fertilizer, which is assumed to be taken from the Norwegian grid for all scenarios, except for the biochar-fertilizer with CHP scenario where it is taken from the electricity output of the pyrolysis plant. Emissions of particulate matter from the grinding and pelletization of the biochar-fertilizer are taken as proxy from the ecoinvent process of lignite briquetting. Emissions of heavy metals associated with the particulate matter are accounted for assuming that the particles are biochar and using heavy metals concentration in biochar as in (Sørmo et al., 2020).

The inventory for the biochar-fertilizer production is shown in Table S6.

2.6. Biochar application to soil

For estimating transport distances for biochar application to the field, each county is assigned one of the conversion plants based on proximity and equally shared grain land area. Distance from the county's capital and conversion plant is considered as a proxy for transportation distances or assumed to be 40 km if biochar is applied to a field within the same county. Distances are weighted by the county's share of grain land area and an average transport distance of 226 km is estimated.

Biochar application to the field is assumed to be broadcasted and followed by harrowing for incorporation into soil. It is assumed that 74% of the carbon in biochar remains in soil after 100 years based on biochar stability in soils measured under Norwegian conditions (Budai et al., 2016). It is assumed that all the calcium in the feedstock remains in biochar as $CaCO_3$, reducing the need for liming by 145 kg year^{-1}. The inventory is available in Table S7.

2.7. Biochar's effects on soil emissions

Emission factors from soils in the reference system are taken from the Norwegian emissions inventory report (Miljødirektoratet, 2019). Soil N_2O emissions from fertilizers are estimated considering that 1% of the nitrogen applied, 1% of the volatized nitrogen and 0.75% of the leached nitrogen are emitted as N_2O. NO_x emissions are 0.04 kg NO_x per kg nitrogen applied, NH_3 emissions are 5% of the nitrogen applied, and 22% of the nitrogen applied as fertilizer is leached from the soil as nitrates. Table S8 in the SI provides a summary of these factors and the range used in the uncertainty analysis.

Modelled effects of biochar include changes in soil N_2O, NO_x and NH_3 emissions and in nitrogen leaching. Direct biochar application to soil in Norway is not expected to have significant effect on grain yield (O'Toole et al., 2018), as also observed in other Nordic countries (Tammeorg et al., 2014a, 2014b). However, biochar-fertilizer has the potential to improve fertilizer efficiency and can therefore induce a positive effect on yields. A literature survey of 10 studies finds that BCFs based on inorganic fertilizer have an average effect on crop yield of 19%, with a standard deviation of 22% (Chew et al., 2020; González et al., 2015; J. Liao et al., 2020; Magrini-Bair et al., 2009; Puga et al., 2020; Qian et al., 2014; Schmidt et al., 2017; Shi et al., 2020; Wen et al., 2017; Yao et al., 2015). An uncertainty range of −3% to +41% for the effects of BCF on grain yields was therefore considered in our analysis.

Given the high uncertainty of effects on soil emissions, uncertainty ranges are considered in a Monte-Carlo analysis. The reduction potential of biochar on N_2O emissions from soils is considered to be between 22 and 50% (with an average effect of 38%), according to a meta-analysis (Borchard et al., 2019). This range is consistent with results from regression modeling for biochar from wood under Norwegian soil conditions under low application rate (0–10 tonnes per hectare) (Liu et al., 2019), and with observed field measures in Norway (O'Toole et al., 2014). Biochar's effect on ammonia volatilization is modeled using regression modeling for biochar from wood under Norwegian soil conditions and low application rate of 0–10 tonnes biochar per hectare (Liu et al., 2019). According to these data, NH_3 volatilization increases between 0 and 10%, with an assumed average increase of 5%. Biochar's effect on soil NO_x emissions from nitrogen fertilizer is based on a review of literature data (Fan et al., 2020, 2017; X. Liao et al., 2020, p.; Nelissen et al., 2014; Niu et al., 2018; Obia et al., 2015; Wang et al., 2019; Weldon et al., 2019; Xiang et al., 2015; Zhang et al., 2019, 2016). NO_x reductions can be as high as 75–80% for biochar produced at high temperature and at high biochar application rates (Wang et al., 2019; Weldon et al., 2019). However, increased NO_x emissions under biochar amendment can also be observed, but mainly from biochar produced at low temperature (<400 °C) (Weldon et al., 2019). At biochar application rates of 3–3.75 tonnes/ha, NO_x reductions of 5–20% are reported (X. Liao et al., 2020; Niu et al., 2018; Xiang et al., 2015). In our scenarios, biochar is produced at 500 °C and an increase in NO_x emissions is not expected. The lower bound of the uncertainty range is thus set at 0%, the average reduction at 10% and the upper bound at 20%. Biochar's effect on nitrogen leaching is taken from (Liu et al., 2019), and it is expected to be a reduction by 0–16% (average 8%). It is assumed that biochar and biochar-fertilizer have the same effect on soil emissions.

Biochar's effects on soil are considered to be effective only for one year after its application, according to recent evidence (Borchard et al., 2019; Liu et al., 2019). It is assumed that biochar is applied annually and long-term effects of biochar on crop yield and nitrogen leaching are not included in the analysis as they are still unclear and uncertain (Borchard et al., 2019; Jeffery et al., 2017).

2.8. Climate and other environmental impacts

The climate impact analysis includes the effects of both greenhouse gasses (CO_2, N_2O and CH_4) and NTCFs (NO_x, CO, SO_x, non-methane volatile organic compounds (NMVOC), organic carbon (OC), black carbon (BC)). These different climate forcers affect the climate system on different time scales: GHGs have long life-time that allows for uniform atmospheric mixing and affect the climate globally; whereas NTCFs have short life-time, are not well-mixed in the atmosphere, and their climate impacts are highly heterogeneous (Levasseur et al., 2016b; Myhre et al., 2013). A single metric like the GWP100 can never capture the full picture of the climate impacts from forcing agents with such a variety of timescales. To overcome these limitations, the United Nations Environment Programme-Society of Environmental Toxicology and Chemistry Life-Cycle Initiative proposed the combined use of multiple metrics that quantify the effects of different climate forcers on different timescales, for example in terms of the rate of climate change or long-term temperature increase (Cherubini et al., 2016; Jolliet et al., 2018; Levasseur et al., 2016a). These metrics are GWP20 and GWP100 to assess short-term and mid-term impacts, and the global temperature change potential (GTP) with TH of 100, GTP100 (Levasseur et al., 2016b). GTP is a metric that evaluates the contribution of an emission to global average temperature at a specific point in time in the future indicated by the TH. A detailed description of these metrics can be found elsewhere (Joos et al., 2013; Myhre et al., 2013; Shine et al., 2005). Since GWP100 characterization factors are numerically similar to the values of GTP40, GWP100 can be interpreted as a metric assessing temperature changes within approximately 40 years (Allen et al., 2016). GWP20 and GWP100 can thus mostly capture short (GWP20) and medium-term (GWP100) climate change impacts that are relevant for the rate of climate change, and, since they are based on integrated (cumulative) effects, they tend to assign relatively higher importance to short-lived forcers like NTCFs or CH_4 (especially for short TH, as in GWP20). GTP100 represents the instantaneous (i.e., non-integrated) effects on temperature at 100 years. It is therefore a proxy for long-term climate impacts and the temperature stabilization goal stated in the Paris Agreement (Levasseur et al., 2016b; Tanaka et al., 2019). In our analysis, GWP20 and GWP100 include the effect of both NTCFs and GHGs, while GTP100 only quantify contributions from GHGs (the ones from NTCFs are negligible).

Characterization factors for NTCFs for GWP20 and GWP100 are taken from (Levasseur et al., 2016b), and are based on world average estimates available from the latest IPCC Assessment Report (Myhre et al., 2013). Values and uncertainty ranges for all the characterization factors are reported in Table S9 in the SI.

We selected six additional impact categories to investigate potential trade-offs or co-benefits: stratospheric ozone depletion, fine particulate matter formation, tropospheric ozone formation, terrestrial acidification, marine eutrophication and terrestrial ecotoxicity. Different types of emissions contribute to varying impact categories. For example, N_2O emissions contribute to stratospheric ozone depletion (in addition to climate change), NO_x participates in tropospheric ozone formation with implication for human and ecosystem health, ammonia (and NO_x) contributes to terrestrial acidification (with potential impacts on plant diversity) and to fine particulate matter formation (with potential impacts on human health), leaching of nitrogen is associated with marine eutrophication, and emissions of heavy metals are key drivers of terrestrial ecotoxicity impacts. All emissions are characterized using averaged mid-point characterization factors from ReCiPe 2016 v1.1 (Huijbregts et al., 2017).

2.9. Large-scale biochar deployment

The biochar potential from forest residues availability is assumed to be applied annually to the grain producing area in Norway, which on average over the period 2010–2020 is about 0.28 Mha (35% of the cultivated area) (SSB, 2020a). From the amount of forest residues available in the counties and the biochar yields of the pyrolysis process described above, we estimate a national biochar production potential of 0.48 ± 0.03 Mtonnes year^{-1}. Assuming an application rate to agricultural soils of 2.5 tonnes year^{-1}, a total of 0.19 ± 0.01 Mha can be annually treated with biochar (representing about 68% of the grain cultivated area). Changes induced by biochar or biochar-fertilizer to barley yields and soil emissions are estimated by considering the specific average effects (and uncertainty ranges) mentioned above over all the treated area.

2.10. Uncertainty analysis

In addition to the uncertainty ranges presented in the previous sections (mostly about soil emissions), our uncertainty analysis considers variability in a range of key parameters that are relevant in the biochar value chain. Uncertainty factors are used for biochar yields, carbon content in biochar and its long-term stability, carbon content in bio-oil, heat required by pyrolysis, transport distances ($\pm$ 20%) of feedstocks or biochar, climate metrics, biochar's effect on crop yield and soil emissions. Biomass composition, such as moisture or ash content, can influence both yield and fixed carbon content of biochar (Peters et al., 2015; Woolf et al., 2014). Variability in biochar yield, carbon content and stability in the uncertainty analysis is performed to capture these variations. Among the uncertainty factors, a key role is played by biochar yields, because it affects emission factors for pyrolysis, the amount of feedstock per kg of biochar to be extracted and transported, and ultimately the total amount of land that can be treated. Further, BC and OC emissions are not included in the emission inventory database, and they are estimated by multiplying PM10 emissions with factors representing the shares of BC and OC emissions from both stationary and mobile sources (Bond et al., 2004). The uncertainty analysis is performed with a comprehensive Monte-Carlo analysis, where 10,000 runs produce results by randomly selecting one value within each of the uncertainty ranges per each run. LCA usually relies on lognormal distribution for uncertainty analysis of parameters, because of qualitative appraisal of knowledge strength using a pedigree matrix approach (Ciroth et al., 2016; Funtowizc and Raveitz, 1990). In our study, we gathered, when available, quantitative literature data on various parameters and establishing a normal distribution was not always possible due to limited sample size. A triangular distribution was thus selected, as recommended by the principle of maximum entropy (Mishra and Datta-Gupta, 2018; van der Spek et al., 2020). The minimum, maximum and mode of each parameters define the triangular distribution. The uncertainty factors and ranges of values is available in Tables S8, S9 and S10 in the SI.

3. Results and discussion

3.1. Climate change impacts

Fig. 2 shows the results (GWP100) for the reference case and the four biochar scenarios considered in our analysis. These results include the effects of both GHGs and NTCFs and show contributions by life-cycle stage (Fig. 2a) or climate forcing agent (Fig. 2b).

In the reference system, managing one hectare of land for barley production without biochar causes about 2.8 ± 0.2 tonnes CO_2eq. ha^{-1} year^{-1}. A key step is fertilizer production (1.13 tonnes CO_2eq. ha^{-1} year^{-1}) followed by farming operation (0.76 tonnes CO_2eq. ha^{-1} year^{-1}). Soil emissions account for 0.67 tonnes CO_2eq. ha^{-1} year^{-1}. There is a similar share of impact from CO_2 and N_2O with 1.23 and 1.42 tonnes CO_2eq. ha^{-1} year^{-1}, respectively. About half of the N_2O emissions in the reference system are due to soil emissions, while the other half comes from nitric acid production for ammonium nitrate supply.

Producing barley in one hectare of land with biochar has a net climate impact of -1.72 ± 0.45 tonnes CO_2eq. ha^{-1} year^{-1}. Farm operations remains the second main contributor to warming emissions, which are higher than those in the reference system (about 85 kg CO_2eq. ha^{-1} year^{-1}) because of additional emissions from biochar application (spreading and harrowing). On the other hand, the reduction in liming use due to biochar reduces emissions by about 76 kg CO_2eq. ha^{-1} year^{-1}. Transportation activities (including both the transport of the feedstock from the forest to the biochar plant and that of biochar from the plant to the field) cause about 0.62 tonnes CO_2eq. ha^{-1} year^{-1}. Pyrolysis does not significantly contribute to direct warming emissions, as power consumption comes from the low-carbon Norwegian electricity grid, which mostly consists of hydropower. Pyrolysis emissions contribute to slightly cooling effects from emissions of NO_x and SO_x. Soil emissions are reduced by about 0.22 tonnes CO_2eq. ha^{-1} year^{-1} compared to the reference case (from 0.67 to 0.45 tonnes CO_2eq. ha^{-1} year^{-1}). Biochar causes both a cooling effect by reducing soil N_2O emissions and a warming effect by reducing soil NO_x emissions (which is a cooling agent), but, because the former is larger than the latter and N_2O has a stronger climate effect than NO_x with GWP100, the net effect is a reduction in characterized emissions. The application of 2.5 tonnes of biochar per hectare also allows the sequestration of 5.35 ± 0.33 tonnes CO_2eq. ha^{-1} year^{-1} in agricultural soils. This amount of negative emissions is larger than the warming effects from emissions along the biochar's value chain and from the farm, so the system has net negative emissions also under a life-cycle perspective. Warming contributions from black carbon and cooling contributions from NO_x and SO_x are increased compared to the reference case, due to the added fuel consumption during the feedstock collection and transportation processes in the biochar supply chain.

Results from the biochar-fertilizer scenario are similar to the biochar scenario. The fertilization stage accounts for the production of the biochar-fertilizer (e.g. grinding and pelletization) and emissions associated with fertilizers production. Power consumption for production of the biochar-fertilizer and higher transport needs due to the increased weight of the biochar loaded with fertilizers are among the key factors for the lower net climate impacts compared to biochar (-1.65 ± 0.48 vs. -1.72 ± 45 tonnes CO_2eq. ha^{-1} year^{-1}).

The biochar-fertilizer with CHP scenario has a climate effect of -4.59 ± 0.74 tonnes CO_2eq. ha^{-1} year^{-1}. Results follow the same pattern of the biochar-fertilizer scenario, but with additional climate benefits from substituting electricity generation and heat production (assumed from natural gas). Avoided emissions are mostly from reducing burning natural gas (96% of the benefits), given the low carbon intensity of the Norwegian electricity mix. The small cooling effect of CH_4 is due to avoided methane losses in the supply chain of natural gas for heat production.

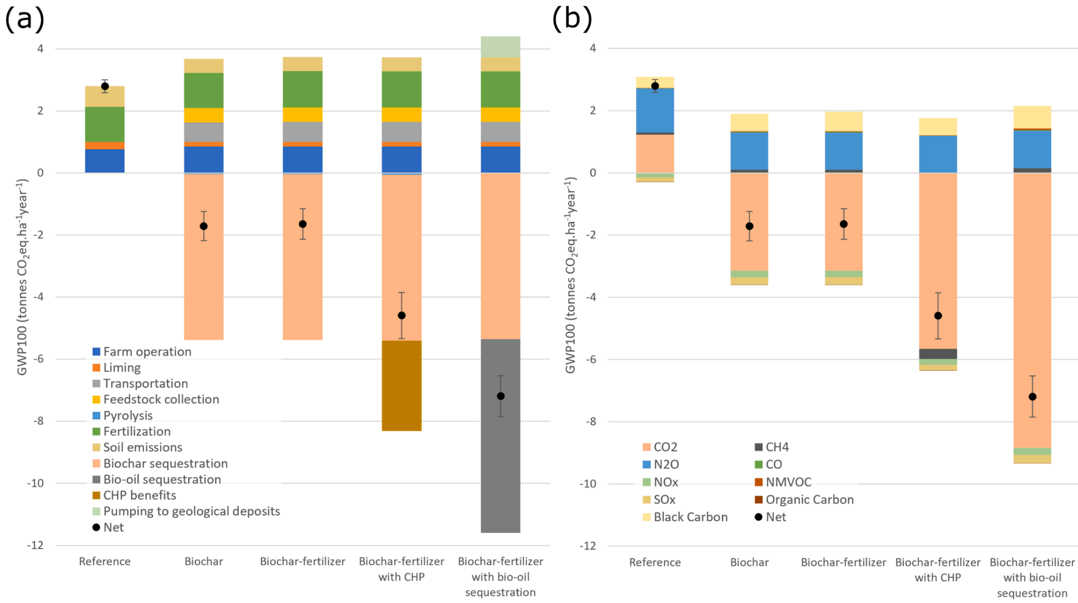

Fig. 2. Climate change effects of the biochar scenarios against a reference system. Results are based on the use of GWP100 to characterize climate impacts and include contributions from both near-term climate forcers (NTCFs) and greenhouse gasses. Both contributions by life-cycle stages (a) and climate forcing agents (b) are shown. Transportation accounts for both feedstock and biochar. Black dots represent the net climate impact and the whiskers show uncertainty ranges from the Monte-Carlo analysis (± one standard deviation).

The biochar-fertilizer with bio-oil sequestration scenario can achieve the largest negative emissions, at −7.19 ± 0.66 tonnes CO_2eq. ha^{-1} $year^{-1}$. Results follow the same pattern as the biochar-fertilizer scenario, but with an additional carbon sequestration from bio-oil of 6.23 ± 0.49 tonnes CO_2eq. ha^{-1} $year^{-1}$. Transport and sequestration of the bio-oil to off-shore geological deposits add 0.69 tonnes CO_2eq. ha^{-1} $year^{-1}$. This means that using excess bio-oil for long-term storage provides larger climate change mitigation benefits than using it to supply heat and power. These results are clearly sensitive to the background energy system, and may vary in other locations where, for example, coal is a primary source for heat or the electricity supply is more dependent on fossil energy sources than Norway.

Figures S6–8 in the SI show the results according to alternative functional units, namely, kg barley, kg biochar and kg feedstock. In terms of impacts per kg barley, the difference between the biochar and biochar-fertilizer scenarios is larger. The climate mitigation is slightly smaller for the latter because BCF increases barley yields, but not biochar production. This implies that the climate mitigation of biochar-fertilizer is spread over a larger grain production and the net benefits are divided by a larger number (as yields are higher), so lowering climate mitigation potential per kg barley as compared to the biochar scenario.

3.2. *Sensitivity of results to climate metrics*

Figs. 3a and 3b show the sensitivity to the use of alternative climate metrics representative of different types of impacts and time perspectives. GWP20, GWP100 and GTP100 are climate metrics that measure the climate system response within a short, mid and long-term period, respectively (see Section 2.8). GWP20 is a metric that focuses on the very short-term and attributes relatively higher importance to NTCFs. It can be interpreted as an indicator to the impact to the rate of climate change. GTP100 is a long-term metric that addresses the temperature stabilization as stated by the Paris Agreements, and it gives comparably little importance to NTCFs and short-lived GHGs (like CH_4). GWP100 lies in between, and it can be interpreted as a metric assessing temperature impacts within about four decades after emissions.

In general, the net climate effects tend to decrease with the longer time perspective of the climate metric (GWP20 – GWP100 – GTP100). This is mainly due to the smaller effect from NTCFs, especially BC, NO_x and CH_4, when a longer TH is considered. For the reference scenario, it means reduced warming, while for the biochar scenarios it means increased cooling. In all the cases, the contributions of the life-cycle stages remain similar across the climate metrics. For the biochar-fertilizer with CHP scenario, the net climate impact remains the same for all climate metrics considered. This occurs essentially because changes in cooling effects are nearly entirely compensated by changes in warming effects.

Warming contributions from soil emissions increase as time perspective increases, because cooling effects of NO_x emissions become less important relative to warming from N_2O (which remains approximately constant) at longer TH. Emissions associated with pyrolysis have larger cooling effects with GWP20 compared to the other metrics due to the higher cooling of NO_x and SO_x at shorter TH, and the impact decreases over longer time scales.

Finally, uncertainty in the climate response decreases as time perspective increases. Uncertainty ranges for GWP20 and GWP100 are dominated by intrinsic uncertainties in characterization factors for NTCFs. These uncertainties are particularly relevant for biochar and biochar-fertilizer scenarios under GWP20, where the ranges are large and the net climate effects can either be of strong cooling or nearly climate neutral (if not slightly positive). For example, characterization factors for BC can range from 270 to 6200 kg CO_2eq. kg^{-1} (mean: 3200 kg CO_2eq. kg^{-1}), or for NO_x from −53 to −27 kg CO_2eq. kg^{-1} (mean: −40 kg CO_2eq. kg^{-1}) (see Table S8 in the SI).

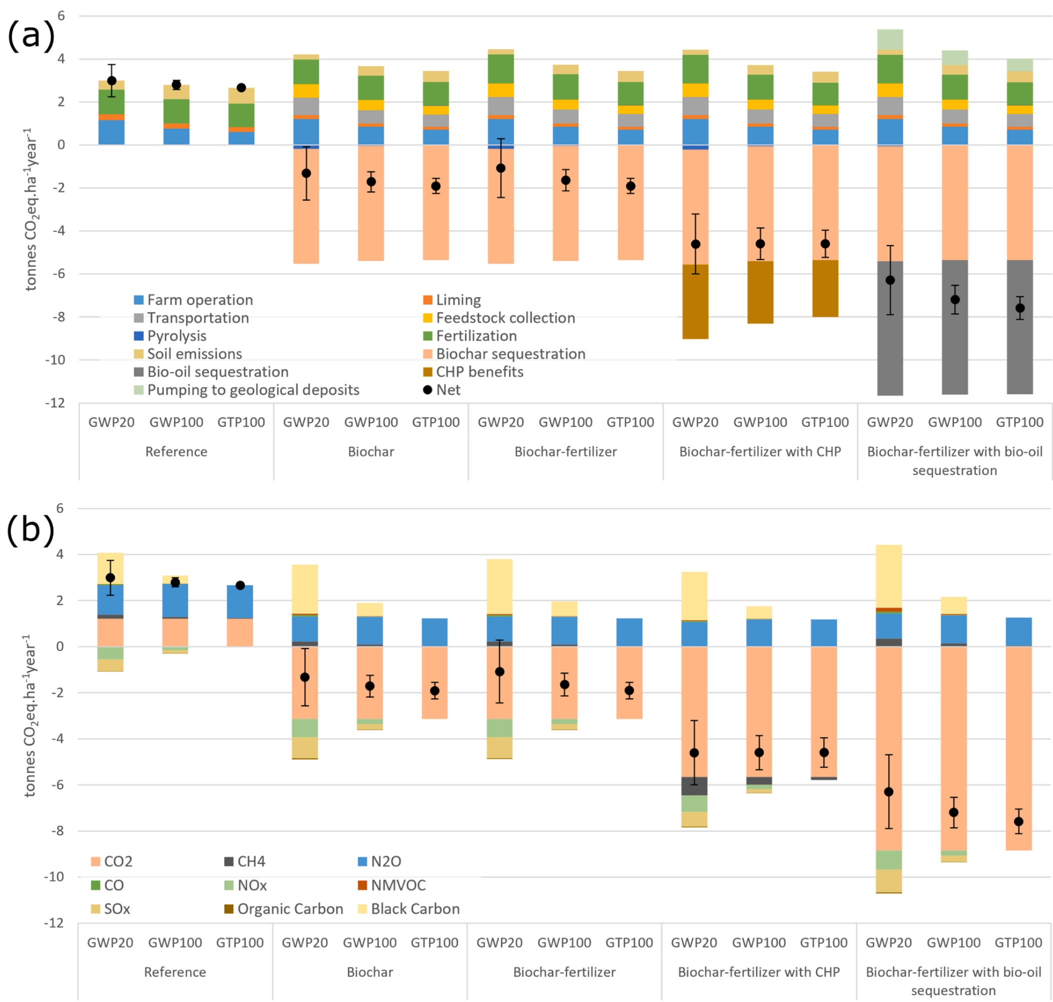

Fig. 3. Climate change effects using different metrics for characterization of impacts: global warming potential at 20 years' time horizon (GWP20), global warming potential at 100 years' time horizon (GWP100) and global temperature potential at 100 years' time horizon (GTP100). Results are presented by life-cycle stage (a) and by contributions of the climate forcing agents (b). Black dots represent the net impact and the whiskers show uncertainty range from our Monte-Carlo analysis (± one standard deviation).

3.3. Net climate mitigation of biochar scenarios

Fig. 4 shows the net mitigation potential of the different biochar scenarios by taking the difference between the climate impact of each given biochar scenario and that of the reference system. In all the cases and irrespective of the climate metric, a net climate mitigation is achieved. Considering each metric and the corresponding uncertainty range, negative emissions can range from −3.7 tonnes CO_2eq. ha^{-1} year^{-1} (GWP20, higher end) to −4.9 tonnes CO_2eq. ha^{-1} year^{-1} (GTP100, lower end) for the simplest biochar scenario, from −3.3 tonnes CO_2eq. ha^{-1} year^{-1} (GWP20, higher end) to −4.9 tonnes CO_2eq. ha^{-1} year^{-1} (GTP100, lower end) for the biochar-fertilizer system, from −6.7 tonnes CO_2eq. ha^{-1} year^{-1} (GTP100, higher end) to −8.5 tonnes CO_2eq. ha^{-1} year^{-1} (GWP20, lower end) for the biochar-fertilizer with CHP and −8.3 tonnes CO_2eq. ha^{-1} year^{-1} (GWP20, higher end) to −10.8 tonnes CO_2eq. ha^{-1} year^{-1} (GTP100, lower end) for the biochar-fertilizer with bio-oil sequestration. Overall, the net mitigation is relatively insensitive to the climate metric used, as all results of each scenario are within the respective uncertainty ranges. In particular, biochar and biochar-fertilizer scenarios have similar net mitigation. If coproducts of the pyrolysis are used to generate heat and electricity, about 65% more climate mitigation is achieved, compared to only producing biochar. Sequestration of the bio-oil into geological deposits can potentially more than double the net climate benefits of biochar alone (+ 120%).

3.4. Other environmental impact categories

Fig. 5 shows an overview of the results for other environmental impact categories of the reference case and the different biochar scenarios. Results are normalized relative to the impact from the reference case in each category. Absolute results are presented in Figures S9-S14 in the SI.

Biochar application to agricultural soils can provide co-benefits in

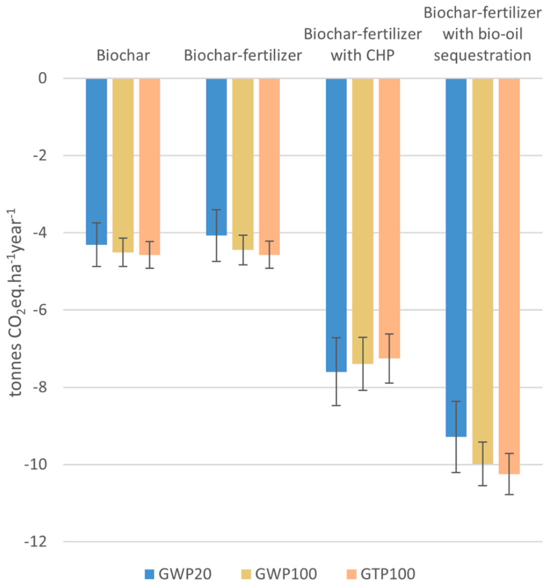

Fig. 4. Net climate change mitigation per biochar scenario and climate metric. Net mitigation is defined as the climate impacts of the given scenario minus the climate impacts of the reference system. Black whiskers show uncertainty ranges from the Monte-Carlo analysis (± one standard deviation).

terms of stratospheric ozone depletion and marine eutrophication, although for the latter the uncertainty range prevent drawing robust conclusions. The magnitude of these co-benefits is relatively insensitive to the type of biochar scenario. On the other hand, tropospheric ozone formation (which affects human health), fine particulate matter formation, terrestrial acidification and terrestrial ecotoxicity have higher impacts for the biochar scenarios than the reference case.

In general, co-benefits occur for those impact categories where biochar's value chain (e.g. transportation, feedstock collection, pyrolysis) does not contribute with relevant emissions. Stratospheric ozone depletion impacts are mainly due to N_2O emissions from nitrogen fertilizers production and soil emissions. Reduction in soil N_2O emissions by biochar explains the lower impacts in stratospheric ozone depletion. Marine eutrophication is mostly driven by soil leaching of nitrogen from the fertilizers, and the biochar's mitigation potential for nitrogen leaching explains the reduced impacts.

In the reference system, contributions to tropospheric ozone formation are mostly due to NO_x and NMVOC emissions from combustion of fuels during land management and soil NO_x emissions from nitrogen fertilizer use. In the different biochar scenarios, there is a reduction in NO_x emissions from soils by 10%, but it is outweighed by higher emissions of NO_x (and to less extent NMVOC) from the combustion of fuels during transportation, feedstock collection and pyrolysis. In the biochar-fertilizer with CHP, avoided production of heat from natural gas prevents some NO_x emissions, which is the reason for the overall lower impacts compared to the other biochar scenarios. In the case of biochar-fertilizer with bio-oil sequestration, the pyrolysis stage has almost no impacts because there are much less NO_x emissions (most of the bio-oil is recovered rather than burnt), but the additional emissions from transportation and sequestration of the bio-oil more than offsets this reduction, and make this scenario the one with the highest impact in tropospheric ozone formation.

Fine particulate matter is mostly formed by emissions of particulate matter (PM2.5) and aerosol precursors like NO_x, NH_3 and SO_x, and it is a potential threat to human health. In the reference system, nearly half of the impact comes from farm operations, and the other half from soil emissions. In terms of individual drivers, the most relevant are SO_x emissions from fertilizers production and emissions of PM2.5 and NO_x from fertilizer production and use of fossil fuels in machineries. Emissions of NH_3 and NO_x from soils lead to the remaining impact for the reference system. Under the biochar scenarios, the combined effect of increase in NH_3 and decrease in NO_x emissions from soils due to biochar application leads to a slight increase in impact from soil emissions of about 2%. This is due to the fact NH_3 is more than twice more impactful compared to NO_x (Huijbregts et al., 2017). The additional emissions of NO_x, SO_x and PM2.5 from combustion processes during transportation, feedstock collection, pyrolysis and biochar-fertilizer production leads to larger impact for all the biochar scenarios compared to the reference case. For the biochar-fertilizer with CHP scenario, there are emissions of NO_x avoided by the displacement of heat from natural gas, leading to the lowest impact score for fine particulate matter formation. For the biochar-fertilizer with bio-oil sequestration scenario, less material is burned in the pyrolysis process with lower emissions of NO_x and SO_x. However, also in this case higher emissions of these compounds during transportation and sequestration of the bio-oil lead to the highest impact in this category.

Deposition of NO_x, SO_x, and NH_3 on terrestrial ecosystems lead to terrestrial acidification, which is a threat to ecosystem health and functioning. Soil emissions are the main contributors to the acidification potential in the reference case and all biochar scenarios and are due to the emissions of NH_3 and NO_x from fertilizer use. About a third of the impact comes from farm operations and mainly from SO_x, NH_3 and NO_x emissions during fertilizer production (due to ammonia and sulfuric acid production) and NO_x emissions from machinery use. In the biochar scenarios, the combined effect of increase in NH_3 and decrease in NO_x emissions from soils due to biochar application lead to a slight increase in impact from soil emissions of about 3% (the acidification potential of ammonia is 5.4 times larger than NO_x (Huijbregts et al., 2017)). Emissions of NO_x and SO_x during pyrolysis contribute to 7% of the impact, while transport and feedstock collection account for 12% together. For the biochar-fertilizer with CHP scenario, avoided use of heat from natural gas saves emissions of NO_x, leading to the lowest impact score for terrestrial acidification among the different biochar scenarios. For the biochar-fertilizer with bio-oil sequestration scenario, lower impact is observed for the pyrolysis process due to less material burnt (and less emissions of NO_x and SO_x), but these savings are more than compensated by higher emissions of these compounds from transportation and sequestration of the bio-oil.

Terrestrial ecotoxicity impacts in the reference system are from emissions of heavy metals during fertilizer production (about 63%, 45% from ammonium nitrate production only), and the remaining are mostly from heavy metals emissions from combustion of fossil fuels in agricultural machinery during farming operation. Contributions of pesticide in soils are below 0.5% of the total impact. The higher needs for transportation of materials and the emissions of pollutants from the pyrolysis stage make the effects on terrestrial ecotoxicity from the biochar scenarios from 3.5 to 4.5 larger than those from the reference system. In the biochar scenarios, transport becomes the main contributor to terrestrial ecotoxicity, with emissions of heavy metals from fossil fuels combustion, mostly copper (92%) and zinc (5%). Biochar-fertilizer production's impacts are largely due to emissions of heavy metals from grinding and pelletization. Impacts from pyrolysis come from emissions of heavy metals (mostly copper 78% and nickel 11%) during combustion of the biochar's co-products. Contribution of PAH emissions during pyrolysis are negligible (lower than 0.0001% of the pyrolysis process's impact). Pyrolysis impacts are lower in the case of bio-oil recovery and sequestration, because it is assumed that most of the heavy metals are recovered with the bio-oil. However, these lower impacts are partly offset by emissions during transport and sequestration processes of the bio-oil.

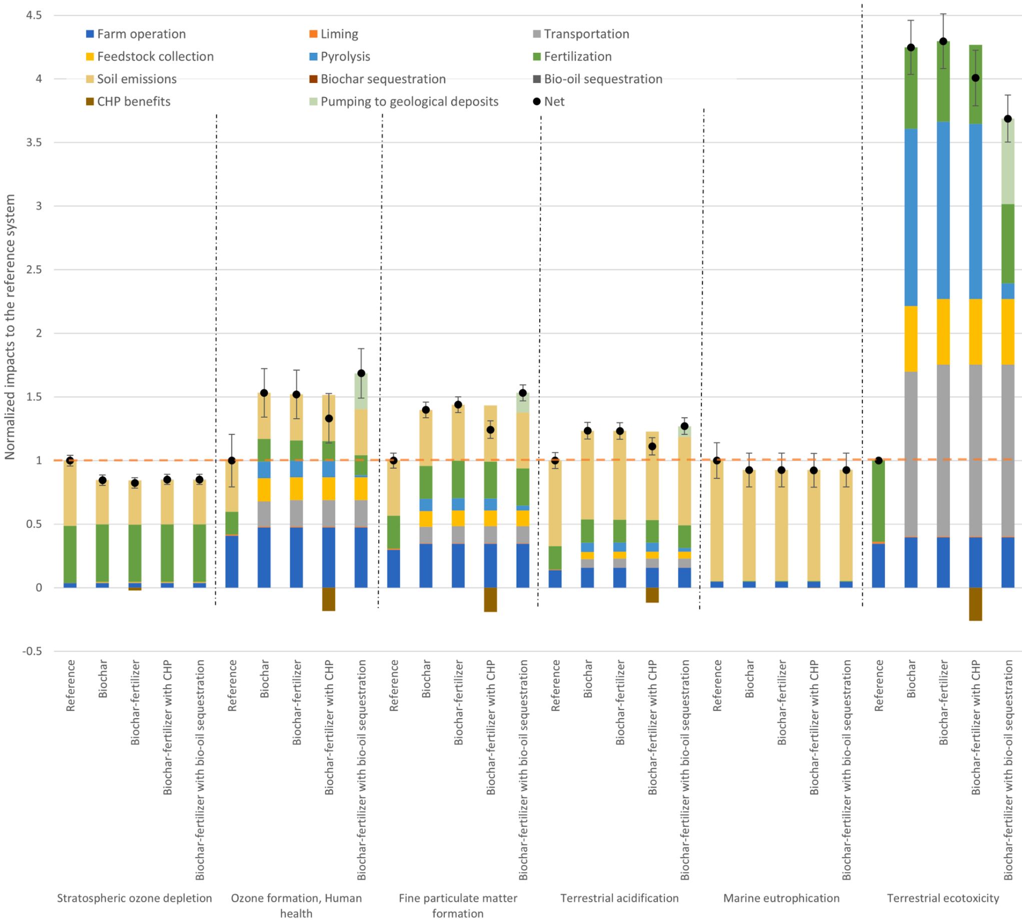

Fig. 5. Life-cycle impacts from the reference system and the four biochar scenarios for 6 impact categories: stratospheric ozone depletion, ozone formation (human health), fine particulate matter formation, terrestrial acidification and terrestrial ecotoxicity. Results are presented by life-cycle stages and are normalized to the impact of the reference system per each category. Transportation accounts for transportation of both feedstock and biochar. Black dots represent the net impact and the whiskers show uncertainty ranges from the Monte-Carlo analysis (± one standard deviation).

3.5. Effects at a national level

Fig. 6 offers an overview of the potential of carbon sequestration (or negative emissions) from a large-scale deployment in Norway of the biochar scenarios analyzed in this study, either with or without a life-cycle perspective. Deployment scenarios are calculated by scaling up the biochar production potential to the total feedstock available, with the associated logistics described in the methods. From the estimated 1.7 Mtonnes of forest residues available per year, about 0.48 ± 0.03 Mtonnes year^{-1}of biochar are produced. Assuming an average application rate to agricultural soils of 2.5 tonnes biochar per ha, about 68% of the 0.28 Mha of grain producing land in Norway can be annually treated with biochar.

Accounting only for the carbon sequestered without a life-cycle perspective, the mitigation potential is 1.01 ± 0.1 Mtonnes CO_2eq. year^{-1}, and it can be about twice as much (2.19 ± 0.1 Mtonnes CO_2eq. year^{-1}) when bio-oil is also captured and stored. Under a life-cycle perspective that accounts for emissions along the whole supply chain, the mitigation potential in the biochar and biochar-fertilizer scenarios is reduced by 15–24%. Adding the generation of electricity and heat adds 36–42% to the climate mitigation of the simple biochar scenario. The consideration of life-cycle emissions in the case of bio-oil sequestration reduces the climate change mitigation potential by 12–20% relative to the case where only the carbon in biochar and bio-oil is taken into account. With the exception of the scenario of biochar-fertilizer with CHP, the life-cycle based yearly mitigation potentials tend to increase when extending the temporal perspective of the climate metric.

Relative to the Norwegian territorial GHG emissions in 2019 (SSB, 2020c), the carbon storage from the biochar without and with bio-oil sequestration can mitigate 2.0% ± 0.2% and 4.3% ± 0.2% of the national emissions, respectively. Taking life-cycle emissions into considerations for the different metrics and uncertainty ranges, the mitigation potential is between 1.3% (biochar-fertilizer, GWP20) and 1.9% (biochar, GTP100) and between 3.1% (biochar-fertilizer with bio-oil sequestration, GWP20) and 4.0% (biochar-fertilizer with bio-oil sequestration, GTP100) respectively. Compared to emissions from the Norwegian agricultural sector only, the climate change mitigation potential of the carbon sequestration in biochar and in biochar and bio-oil is 20.6% ± 1.7% and 44.5% ± 2.1% respectively. Under a life-cycle perspective for the different metrics and uncertainty ranges, these

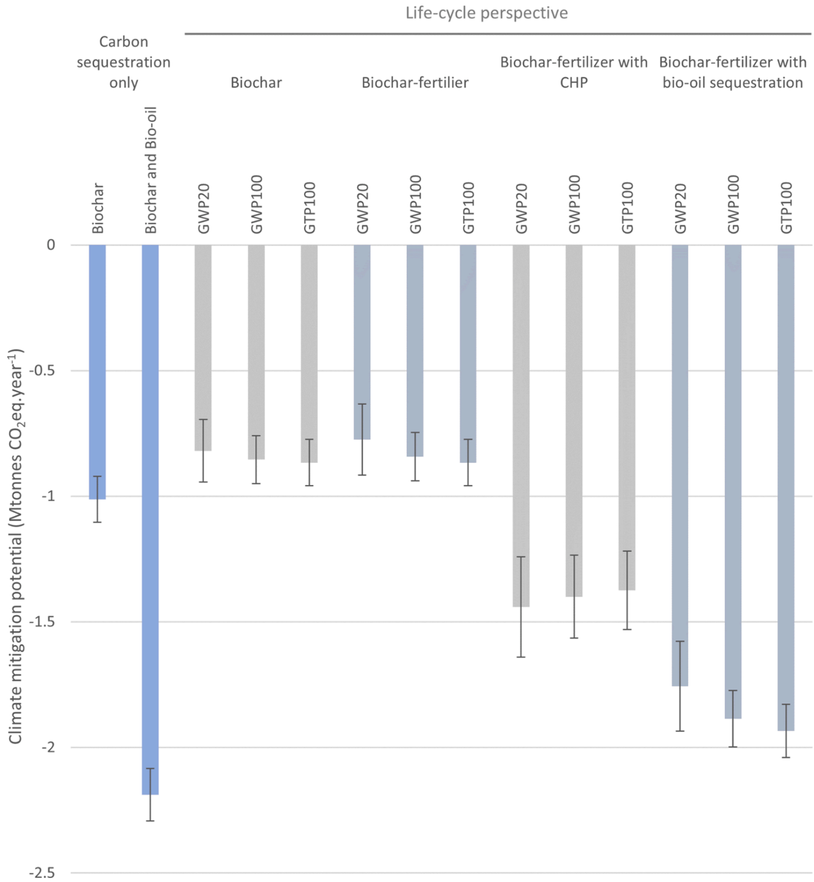

Fig. 6. Comparison of the climate change mitigation potential of a large-scale deployment of biochar in Norway considering only the carbon contained in biochar and bio-oil or taking a life-cycle perspective. Black whiskers show uncertainty ranges from the Monte-Carlo analysis (± one standard deviation).

figures are between 12.9% (biochar-fertilizer, GWP20) and 19.4% (biochar, GTP100) and between 32% (biochar-fertilizer with bio-oil sequestration, GWP20) and 41.4% (biochar-fertilizer with bio-oil sequestration, GTP100).

Fig. 7 shows how the large-scale deployment of biochar in Norwegian agriculture affects yields of barley and soil emissions. Based on national availability of forest residues, biochar can be annually applied to about 0.19 ± 0.1 Mha of grain production area, resulting in a yield increase of about 0.14 ± 0.06 Mtonnes per year (+12%) (under the assumption that all the land is dedicated to barley production). The mitigation of N_2O emissions is 21% ± 4% compared to baseline emissions where land is not treated with biochar. This mitigation is due to a reduction of direct emissions of N_2O from fertilizer application (25% ± 4%), a decrease of indirect N_2O emissions due to a decrease of nitrogen leaching from soils (5% ± 2%), and an increase of about 3% ± 1% of indirect N_2O emissions from the overall increase of ammonia volatilization. Compared to the national statistics for 2019, the reductions of N_2O emissions correspond to 1.8% of the national N_2O emissions and 2.4% of the agricultural N_2O emissions (SSB, 2020d).

The application of biochar causes additional ammonia volatilization by around 3% ± 1%, corresponding to an increase of 0.26% and 0.27% of the national and agricultural total ammonia emissions, respectively (Miljødirektoratet, 2019; SSB, 2020e). The low contributions to both national and agricultural emissions is due to the comparatively high emissions of ammonia from handling of manures from livestock systems.

Soil emissions of NO_x decrease by about 7% ± 3%, corresponding to 1.6% of NO_x emissions from the agricultural sector in Norway. At the total national level, this reduction becomes negligible because agricultural NO_x emissions only represent 5% of the Norwegian emissions (which are dominated by oil and gas extraction and transportation) (SSB, 2020e). The somewhat larger uncertainty range for NO_x emissions from soils comes from the large uncertainty of NO_x emission factor from fertilizer, which can range from 0.005 to 0.104 kg NO_x kg^{-1} N applied (12.5 to 260% of the average emission factor of 0.04 kg NO_x kg^{-1} N applied) (Miljødirektoratet, 2019).

Biochar can reduce nitrogen leaching in agricultural soils by about 5% ± 2%, corresponding to 0.4% of the total anthropogenic nitrogen input to Norwegian coastline or 1.5% of the agricultural nitrogen losses compared to 2018 emissions (Selvik and Sample, 2018). However, uncertainty ranges are large and overlapping.

The potential energy recovery from pyrolysis can produce additional electricity and heat. The electricity potential is 880 ± 180 GWh $year^{-1}$, and heat potential is 1800 ± 370 GWh $year^{-1}$. This electricity generation represents about 0.6% of the electricity production in Norway in 2020 (SSB, 2020f), but heat production from pyrolysis has a larger potential contribution to the national energy system, as it can deliver about 30% of the current district heating production (SSB, 2020 g).

In general, the main co-benefits with climate change mitigation are

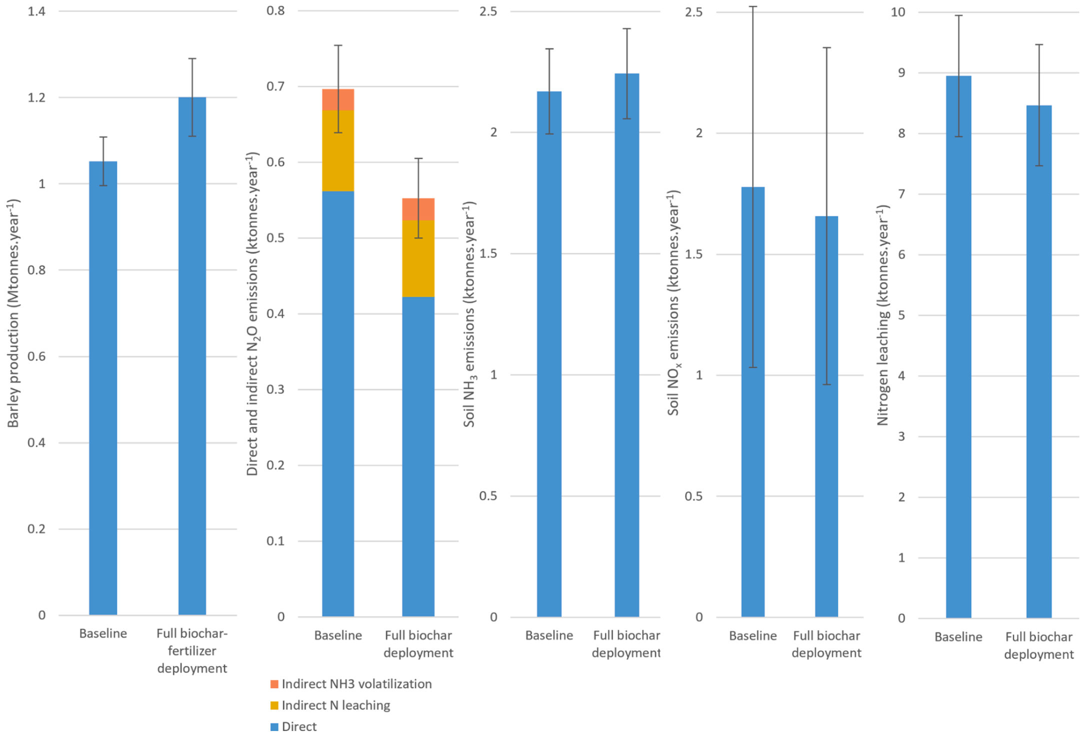

Fig. 7. Effects of a large-scale deployment of biochar application to Norwegian agricultural soils on grain yields, soil emissions (N_2O, NH_3, NO_x) and nitrogen leaching. Black whiskers show uncertainty range from the Monte-Carlo analysis (± one standard deviation). Results only refer to biochar effects in soils, and do not consider life-cycle emissions.

related to increased yields and reduction of N_2O emissions, while the decrease in soil NO_x emissions and nitrogen leaching has less confidence because of the large overlap of the uncertainty ranges. The increase in soil ammonia emissions is also not significant.

4. Uncertainties and limitations

Our results are subject to a series of uncertainties and limitations. In particular, soil conditions, climate and land management affect the level of soil emissions and how biochar alters them. Biochar's production and supply chain are also subject to uncertainties, such as variability in feedstock composition, which can affect yield and biochar's carbon content, or the distances between forest, biochar production plants and fields. The climate response to emissions of NTCFs is also dependent on a variety of factors. Variability on these parameters have been included in the Monte-Carlo analysis to investigate how they influence our results. The results are generally robust to these uncertainty factors, especially climate change effects based on GWP100 and most of the other impact categories (except marine eutrophication). Net negative climate effects from biochar with GWP20 are uncertain in two scenarios, for which the negative emissions of the biochar scenarios can be questioned, but overall a net mitigation relative to the reference system is achieved.

LCA studies are subject to different assumptions regarding system boundaries and other methodological aspects that make results specific to the individual case and comparison across studies challenging (Matuštík et al., 2020; Tisserant and Cherubini, 2019). Our work is specific to the Norwegian context in terms of resource supply, conversion processes, agricultural operations, and soil emissions. This country specific approach offers an estimate of the national potentials, co-benefits and trade-offs associated with alternative biochar utilization scenarios in Norway. Different local conditions and assumptions will evidently lead to different outcomes. For example, countries with a larger fraction of fossil-based electricity in their power mix will show higher avoided emissions in the CHP scenario, or larger application rates of biochar would cause larger effect on soil emissions. Compared to other LCA studies, our results are broadly consistent despite the large variability found in the literature. A recent review that summarized LCA studies of biochar systems with production and application to agricultural fields estimated total climate change impacts of -0.9 ± 0.3 tonnes CO2eq. tonnes^{-1} feedstock (median ± one quartile), but with 5th and 95th percentile of -0.1 and -1.5 tonnes CO2eq. tonnes^{-1} feedstock, respectively (Tisserant and Cherubini, 2019). For comparison, net climate change effects in our study range from -0.25 tonnes CO2eq. tonnes^{-1} feedstock (biochar and biochar-fertilizer scenarios, GWP100) to -0.8 tonnes CO2eq. tonnes^{-1} feedstock (biochar with bio-oil sequestration, GWP100) (Figure S8 in the SI). In line with our analysis, the review found that climate impacts from the biochar value chain can be up to $+0.5$ tonnes CO2eq. tonnes^{-1} feedstock, and those of biochar sequestration in agricultural soil can contribute -0.25 to -0.75 tonnes CO2eq. tonnes^{-1} feedstock (5th and 95th percentile) (we estimated about -0.6 tonnes CO2eq. tonnes^{-1} feedstock in our study).

Quantification of the effects of different deployment scenarios is based around national average soil conditions, fertilization and crop yields. This is a simplification as agricultural production is very heterogeneous and depends on local climate and practices. To take this variability into consideration, Norway's average barley yield over 10 years are used to partially even out the regional differences in management and fluctuations in weather conditions.

The effect of biochar on soil N_2O emissions is also uncertain. Both carbonization degree of the biochar and soil type appear as key factors controlling effects on N_2O emissions. For example a study reports that well carbonized biochar products, i.e., produced at higher temperature, consistently reduced N_2O emissions from two contrasted soil types (Weldon et al., 2019). Less carbonized biochars suppressed N_2O emissions only in a mineral soil but induced the opposite effect in a peat soil. The duration of the effect on N_2O emissions is also a source of variability. A recent biochar review reports an average reduction of N_2O emissions by 38%, but also indicates that reductions tend to be negligible after one year (Borchard et al., 2019). However, these emission reductions can be sustained over time by annual applications of well-carbonized biochar or BCF. In our Monte Carlo analysis, a range of 22 to 50% reduction is considered to take the variability of this effect into account. Although variable, it is important to consider this positive effect of biochar, especially in light of alternate solutions. On average, agronomic practices aiming at increasing carbon sequestration in soil lead to a slight increase in N_2O emissions, while biochar leads to a reduction (Guenet et al., 2021).

In addition to the uncertainties above, there are a range of processes and considerations that have not been investigated in our study. The economic dimension was not explored but it is a necessary component for a successful large-scale deployment of biochar systems. In general, a pyrolysis system with biochar production has a positive net present value at a feeding rate above 9 tonnes per hour (about 45 MW capacity or larger) at pyrolysis temperature above 450 °C, or above 6 tonnes/hour (about 30 MW capacity or larger) at pyrolysis temperature above 550 °C (Yang et al., 2021). An integrated strategy of producing both biochar and bioenergy is found to have higher net present value than simple bioenergy systems, in particular if there are positive effect of biochar on yields (Woolf et al., 2016). At about 290 MW and a pyrolysis temperature of 500 °C, our modelled scenarios for biochar production are thus within these economic viability criteria based on carbon market, bioenergy and biochar prices.

The possible effect of biochar on the degradation rate of native soil carbon stocks, an effect referred to as priming, has not been included in the analysis. On average, the addition of biochar amendments into soil has been reported to decrease the decomposition rate of the native soil organic matter and thereby further increase carbon sequestration (Ding et al., 2018; Wang et al., 2016). However, there is a large variability in these results (Ding et al., 2018; Wang et al., 2016). Low temperature chars are still rich in labile compounds and can increase mineralization, while higher temperature chars reduce the decomposition of organic matter (Chen et al., 2021). In addition, the priming effect of biochar on soil organic matter is often transient (Budai et al., 2016), and the long-term effects, if any, are uncertain.

Reduction of surface albedo due to darkening of soils after biochar application has been suggested to cause a warming feedback that can reduce the climate mitigation potential of biochar (Bozzi et al., 2015; Genesio et al., 2012; Meyer et al., 2012; Verheijen et al., 2013). For example, a study estimates that changes in albedo could reduce climate mitigation of biochar by 13–22% (Meyer et al., 2012). However, this effect is expected to be limited in Norway because of snow cover and low insulation during winter months. At high latitudes, the exposure of a darker soil would be limited to a few weeks in spring between snow melt and crop growth and in the autumn between harvest and snow fall. Further, the second year following a biochar application of 30–60 tonnes.ha^{-1} a decrease in the effect of biochar on soil albedo was observed due to further soil mixing under subsequent tillage operations, thereby reducing the potential changes in surface albedo in cases of one-off applications (Genesio et al., 2012). Albedo changes after biochar application could also be managed by maintaining a canopy cover in between cropping cycle using cover crops, with potential additional benefits in terms of soil carbon accumulation (Jian et al., 2020).

Our analysis does not include the alternative oxidation rate of forest residues left in the forest to decompose (Guest et al., 2013; Ortiz et al., 2014). Part of the residues will become CO_2 in a few years and a smaller fraction will return to soil and litter. The potential inclusion of these fluxes would alter the profile of our results especially in the short term, as residues would represent a sort of temporary short-term carbon storage, but in the long term the residues will largely oxidize to CO_2 in any case. Their collection and use as biochar will move the temporary storage from the forest to the agricultural soils, as part of the carbon in the feedstock goes to biochar. Several reviews and meta-analysis investigate the consequences of removing forest residues after tree harvest on forest productivity, soil nutrient content, soil carbon stock and soil properties with some contrasting results (Achat et al., 2015a, 2015b; Clarke et al., 2021; Hume et al., 2018; Ranius et al., 2018; Wan et al., 2018), in particular for forest soil carbon stocks (Achat et al., 2015b; Hume et al., 2018; Ranius et al., 2018; Wan et al., 2018) and in cold climates (Achat et al., 2015b; Clarke et al., 2021). In general, the level of residues harvested is an important determinant of the effects on forest ecosystems. In our analysis, a conservative extraction rate of 35% is used, which is below the 50% limit recommended for sustainability criteria in nearby Scandinavian countries (de Jong et al., 2017).

Dust emissions from biochar handling, especially during its application, have raised concerns for implications on human health and climate (Gelardi et al., 2019; Genesio et al., 2016). However, these emissions are hard to measure and robust estimates are not readily available in the literature. A BC emission in the range of 0.3–6.7 kg ha^{-1} (0.01–0.26% of 2.5 tonnes ha^{-1} application rate) of biochar dust can result in no net negative emissions for the biochar and biochar-fertilizer scenarios, when short-term climate change impacts are assessed using the uncertainty ranges of the characterization factors for BC with GWP20 (270–6200 kg CO_2eq. kg^{-1}). Options to limit the potential emissions of dust exist, for example by applying the biochar wet and under low wind conditions, or use biochar pellets (as it is in our biochar-fertilizer scenarios) (Gelardi et al., 2019).

Biochar has been shown to reduce availability of heavy metals in soils and limit their uptake by crops (Chen et al., 2018; Hilber et al., 2017), as well as affecting pesticides' fate in soils (Liu et al., 2018). Biochar's effect on heavy metals was not included in our analysis due to a lack of wide-spread data on concentration and availability of heavy metals in Norwegian agricultural soils. Limited data are also available for the effects of biochar on pesticides, and contrasting findings are sometimes reported, with usually lower availability under biochar amendment but mixed effect on their degradation (Liu et al., 2018). Both effects can be potential co-benefits of biochar application to agricultural soils for human health and terrestrial ecotoxicity, but are expected to have little overall influence on our results. It is unlikely that reduction in availability of heavy metals in soils can offset the effect on terrestrial ecotoxicity, which is primarily linked to the emissions of heavy metals in the supply chain of biochar, while pesticides had a negligeable contribution to terrestrial ecotoxicity impacts.

Storage of the bio-oil in geological deposits can have technical challenges and limitations that are to be overcome. Bio-oils are known to have higher viscosity than heavy oil, and the corrosivity can make transport and pumping difficult. However, a review study argues that bio-oils and fossil crude oils have similar properties in terms of pumping and transportation (Schmidt et al., 2018). Bio-oils are also slightly corrosive due to low pH and should be carefully stored, particularly as they contain toxic compounds (Cordella et al., 2012). If geological sequestration of bio-oils turns out unfeasible or uneconomical, bio-oil can be used in a variety of products to replace fossils (for fuels or chemicals) (Pinheiro Pires et al., 2019). Incorporation of bio-oils in asphalt paving would correspond to an alternative form of carbon sequestration.

5. Conclusions

Biochar production is a mature process and one of the most cost-efficient NETs, and can be a strategic option to be developed in the near-term before other technologies emerge. Our analysis shows that negative emissions can be achieved for all scenarios when accounting for a wide range of emissions (both GHGs and NTCFs) along the entire life-cycle. The exclusion of life-cycle emissions leads to an overestimate of the mitigation potential of 10–20% when benefits of co-products are excluded. Including a variety of biochar-induced soil effects in the analysis allowed to quantify potential co-benefits or trade-offs regarding other environmental impact categories: increased food production, reduced stratospheric ozone depletion and, though uncertain, marine eutrophication, while impacts on tropospheric ozone formation, terrestrial acidification, fine particulate matter formation and terrestrial ecotoxicity are increased. Biochar could significantly reduce emissions of N_2O at the Norwegian level, while application of biochar-fertilizer could represent a benefit in terms of increased grain production. However, the effect is more uncertain in terms of reduction of NO_x emission and leaching of nitrogen and increased NH_3 emissions. Integrating emissions from both the supply chain and soils is important to prevent spill-over effects, as we found that some co-benefits in terms of soil emission reduction can be outweighed by emissions happening in the supply chain. Greener future transportation systems and stricter emission control measures at the pyrolysis facilities can mitigate these adverse effects, with additional benefits for tropospheric ozone formation and fine particulate matter formation. These results show the need of taking a holistic approach in terms of accounting emissions along the biochar supply chain and assessing environmental impacts using multiple assessment methods. Better knowledge regarding soil effects can help to guide an optimal management of biochar and agricultural land based on local conditions.

CRediT authorship contribution statement

Alexandre Tisserant: Investigation, Formal analysis, Methodology, Writing – original draft. **Marjorie Morales:** Methodology, Writing – review & editing. **Otavio Cavalett:** Methodology, Writing – review & editing. **Adam O'Toole:** Investigation, Writing – review & editing. **Simon Weldon:** Investigation, Writing – review & editing. **Daniel P. Rasse:** Project administration, Funding acquisition, Writing – review & editing. **Francesco Cherubini:** Conceptualization, Project administration, Funding acquisition, Supervision, Writing – review & editing.

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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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2021.106030.

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Chapter 4. Biochar and its potential to deliver negative emissions and better soil quality in Europe

Submitted to Earth's Future

This paper is submitted for publication and is therefore not included.

Chapter 5. Discussion and conclusion

Chapter 5. Discussion and conclusion

This chapter aim at summarizing the findings of this body of work, further discuss additional points and contrast different aspects that have been investigated, and uncertainties, before concluding.

5.1 Climate change mitigation potential

Taking a life-cycle perspective, biochar can compensate around 1.3% of Norwegian GHG emissions per year, or between 1.6% and 3.7% of European emissions per year depending on feedstock supply scenarios. Energy production from a CHP cycle coupled with the pyrolysis process can offset additional emissions from the electricity grids and natural gas heat, leading to a climate mitigation potential of about 2.3% and between 2.8% and 6.3%. Sequestration of both biochar and co-produced bio-oil can compensate about 2.7% of Norwegian GHG emission and between 3.0% and 6.9% for Europe. In Europe, life-cycle emissions offset about 15% of the carbon sequestration potential of biochar and bio-oil. Large transport distances for bio-oil, assumed to be with road freight, are the main contributor for this offset. Transporting bio-oil using train freight instead would allow to achieve higher level of negative emissions in this case.

Future scenarios from Integrated Assessment Models (IAMs) estimate median values of global cumulative negative emissions (NE) need in 2050 of 84 $GtCO_2eq$ (15 and 189 GtCO2eq as 5 and 95% percentiles), 97 (29 - 327) $GtCO_2eq$ and 121 (12 - 318) $GtCO_2eq$ for a climate change stabilization target at 1.5°C without overshoot, 1.5°C with low overshoot, and 1.5°C with high overshoot, respectively[1]. Comparing with our European scenarios, cumulative emissions over 30 years can achieve in the high residue supply case 5.6, 9.4 and 10.3% of the median cumulative NE for 1.5°C without overshoot, 4.8, 8.2 and 8.8% of the median cumulative NE for 1.5°C with low overshoot and 3.9, 6.6 and 7.2% of the median cumulative NE for 1.5°C with high overshoot. Mobilizing forest and crop residues in Europe over 30 years could provide up to a tenth of estimated global need for NE.

5.2 Large vs. small scale biochar production

Typically economies of scale helps to reduce biochar's cost[2,3]. Large-scale production can allow easier emission control of pyrolysis[4], and on-site collection of large volume of bio-oil, that could be further sequestered or used for energy production (CHP). Large-scale biochar production could also be integrated into biorefinery where pyrolysis co-products can be upgraded to liquid biofuel or other products that could replace fossil-based chemicals[5]. However, cost analysis, energy requirements and consequences for LCA emissions for converting bio-oil into liquid biofuels need to be assessed. Large-scale production requires collecting biochar feedstock over larger areas, therefore requiring more transportation, but I find that feedstock transport offset only a very small proportion of the carbon sequestration of biochar.

Depending on the residues supply potential considered in Europe I find that between 230 and 570 biochar conversion plants are required to treat all the available biomass. Each plant was to convert about 560 ktonnes feedstock, which is large compared to current planned or in operation bio-energy plants in Europe that typically treat up to 250-350 ktonnes residues[6]. This result highlights the level of investments that would be required to deploy biochar at the continental scale, only using available residues.

Of course, large scale biochar conversion plants are not the only solution that can be considered. Small scale pyrolysis could be adapted for farmers that want to produce their own biochar, but could represent a significant investment for individual farmers or cooperatives of farmers. The question is also of whether valorization co-products of pyrolysis can be profitable under small or farmer scale biochar production. Farmers could use heat produced by pyrolysis for animal husbandary[7], farm dwellings[8] or drying grain[9], but feasibility need to be assessed on a per case basis. Bio-oil could potentially be recovered and sold, but careful attention should be paid to its handling as it can be corrosive and contains toxic compounds[10,11].

5.3 Wood- vs. crop-based biochar

Crop residues are readily available to the farmer, which could lower the cost of feedstock procurement if they can invest in knowledge and capital to produce their own biochar or grouped in cooperatives. On the other hand, woody residues are typically more scattered in the landscape, leading to more transportation, while they are typically not directly available to farmers and would represent a net cost.

In the scenarios of biochar deployment in Europe, I find that crop-based biochars have larger potential in stimulating crop yield, reduce soil N_2O emissions compared to wood-based biochar. However, crop-biochar tends to increase NH_3 volatilization in nearly all European soils, whereas wood-based biochar can mitigate it depending on locations and application rates. Treating biochar with acid to remove its liming effect may help mitigating increased soil NH_3 emissions[12,13], but consequences of such treatment on other effect of biochar in soil would need to be studied as well.

5.4 Low vs. large application rate of biochar

Higher application rates (30 t ha^{-1} vs. 5 t ha^{-1}) of biochar can achieve higher mitigation of N_2O, NO_x emissions and nitrogen leaching and higher increase in soil water retention across Europe, but not for crop yield and NH_3. In particular, biochar applied at 30 t ha^{-1} can achieve net mitigation of soil N_2O emissions around the Mediterranean Sea, but not at 5 t ha^{-1}. And wood-based biochar can mitigate NH_3 emissions at 30 t ha^{-1} in almost every European soils, but has mixed effect at 5 t ha^{-1} depending on local soil conditions. Application rate of biochar can be chosen to achieve most benefits in given locations, but will also depend on the investment level that the farmer can afford, as to treat the same amount of land 30 t ha^{-1} would cost 6 times more than 5 t ha^{-1} in purchasing biochar.

Changes in soil emissions and associated impacts in European agriculture depends on the amount of cropland that can be treated per year, which is a function of the amount of biochar available and its application rate. Taking these aspects into consideration, I find that applying biochar at 5 t ha^{-1} can achieve larger mitigation or increase in soil emissions compared to 30 t ha^{-1} because 6 times more land can be treated at 5 compared to 30 t ha^{-1}.

5.5 Direct effect vs. LCA perspective

Both for Norway and for Europe, we find that biochar systems achieve net climate mitigation across various climate metrics, even accounting for emissions of GHGs and NTCFs along the supply chain. However, regarding other impacts categories, I find that biochar's effect on soil can mitigate agricultural impacts on stratospheric ozone depletion, marine eutrophication and tropospheric ozone formation, but increase impacts in terrestrial acidification and fine particulate formation. Increases in those two impacts are driven by the increase in NH_3 emissions from soils. Biochar prepared from crop residues are the main driver for these increases due to its larger availability and overall higher potential to increase NH_3 emissions compared to wood-based biochar. However, the picture changes when accounting also for supply chain emissions. Then, biochar systems consistently only mitigate impacts on stratospheric ozone depletion, marine eutrophication, but not in other impact categories. The largest trade-off in terms of environmental impacts of biochar systems is regarding terrestrial ecotoxicity, where transportation and pyrolysis are two main contributors. Switching to cleaner fuel for transportation could help mitigate this trade-off. For pyrolysis, the impact come from heavy metals bound to fine particulate matter released during the combustion of biochar's co-products. Installing filtration system to remove this particulate matter would allow to reduce pyrolysis contribution to this impact category but also to fine particulate matter formation[4], but was not accounted for in this work.

Biochar production coupled with CHP can provide more co-benefits in terms of environmental impacts, but is highly dependent on the kind of energy it offset. Norway's electricity mix comes from renewables at 98%[14], and biochar production coupled with CHP has lower impacts for all categories compared to the other biochar systems, but not compared to the baseline scenario. On the other hand, 36% of European electricity production comes from fossil fuels[14], offsetting some of this production can achieve net negative impacts for tropospheric ozone formation, terrestrial acidification and fine particulate matter formation. However, as cleaning of European electricity mix in the future is expected to mitigate climate change, it will also reduce these co-benefits.

5.6 Uncertainty and future work

In this work, biochar's effect on soils were only considered for the year following its application. Long-term effects of biochar on soils are still largely debated, though some indications of long-term effects exist in the literature. Based on a meta-analysis, biochar may have only transient effects on soil N_2O emissions, disappearing after one year[15]. Biochar's effect on soil NO_x emissions is less studied N_2O, and its long-term effect have not been investigated. As soil NO_x emissions are usually correlated to N_2O, the effect may be also only transient[16]. Biochar can increase NH_3 volatilization due to its alkalinity, moving chemical equilibrium towards NH_3, while its surface chemistry can help retain NH_4^+ and help reduce its volatilization as NH_3[12]. For these reasons, the increase in NH_3 can be expected to be transient, as oxidation of its surface will help to better retain NH_4^+ and its alkalinity will decline[12]. However, biochar is also advocated to reduce soil bulk density and compaction, thus increasing soil aeration. In this case, the increase in NH_3 volatilization due to biochar could be sustained, if diffusion is the limiting factor to NH_3 volatilization. Biochar can retain nitrate in soils, but it is unclear as how it will develop over years once the mixture of soil and biochar reaches its new maximal capacity. It will likely depend on nitrogen cycling in soil, uptake by plants and input of nitrogen fertilizer. Regarding biochar's effect on soil physical properties, A study noted that the increase in soil water retention was only temporary and faded after 7 years. They suggested that the fading may be due to constant mixing of the soil during tilling and downward migration of biochar over the year[17]. This vertical migration of biochar in the soil profile may particularly happen in coarse soils, which are also the ones most benefiting from biochar for increased soil water retention. For example, downward migration of biochar can be as much of 15-20% below its application layer (i.e 0-10cm) after one year[18]. Regular application of lower load of biochar could help maintain more biochar in the root layer of plants and help provide more sustained benefits to crop. But as with more long-term data on soil effects, investigating different strategies for applying biochar (i.e. single vs. several application) need to be further studies to identified potential co-benefits or trade-offs.

Biochar may also have other effect on carbon sequestration and local climate. One aspect that was not included is its potential to allow for stabilizing and help sequestering additional soil carbon. For example, a study applied 9 t ha^{-1} biochar and found that it stabilized native soil carbon and allowed carbon accumulation, which doubled the carbon sequestration present in the biochar[19]. Neutral and more alkaline soils such as found in Europe, and biochar produced at temperature higher than 500 °C (as considered in this work) are two important control factors that improve this effect[3,20]. Contrary to biochar, which carbon sequestration increases with the amount of feedstock that can be recovered and applied to soils, this effect would scale with the amount of land treated, moderate application rate of biochar rather than high application rate would take more advantage of this effect (i.e. treating more land at a given biochar production potential). Further study may be required to clearly identified the potential and stability of this carbon sequestration due to biochar.

Biochar can also affect local climate by modifying land energy balance, by changing soil albedo and by increasing soil water retention. Reduction of surface albedo due to darkening of soils after biochar application has been suggested to cause a warming feedback that can reduce the climate mitigation potential of biochar[21,22]. Decrease in albedo follows a power decay function with increasing biochar application rate[23], so this warming effect increases with biochar application rate before reaching a plateau. On the other hand, carbon sequestration per unit of land increases linearly with biochar application rate, so does the associated cooling effect. The question then is whether there can be a threshold biochar application rate where albedo’s warming effect can completely offset the carbon sequestration effect. This albedo will vary greatly depending on the level of insulation (i.e. stronger at lower latitude) and the amount of time where soil is exposed without canopy cover. An important sensitivity would be harvesting time, early harvest in summer would leave the soil bare for the summer month when insulation is highest, for example in France harvest of winter wheat, barley or rapeseed can be harvested as early as June[24]. However, this effect may only be transient due to further mixing of the biochar in the soil during the next growing seasons[21]. Albedo changes after biochar application could also be managed by maintaining a canopy cover in between cropping cycle using cover crops or by delaying post-harvest tilling[25]. Biochar’s increasing in soil water retention can allow more water for evapo-transpiration which would have a local cooling effect. Investigating those effects on local climate requires development of global models or including biochar into regional climate model to quantify the relative importance of those two distinct effects.

In this work, climate mitigation potentials of biochar systems were investigated using different climate metrics and quantifying a larger range of emissions than usually performed in LCA footprinting that typically focuses only on GHGs. We noted that there were no trade-offs between short and long-term climate mitigation. However, a potentially relevant emissions for short-term global warming was not properly estimated, which is the potential climate effect of the emissions and atmospheric transport of biochar fine particles during its application and from the field. These particles have been argued to have similar light absorbance capacity as black carbon emissions that have a strong, short-lived warming effect. However, measurement of the visible light absorbance of these particles, such as mass absorption cross section of at least 5 m^2g^{-1} at a wavelength of 550 nm, are still to be done to estimate the climate effect of these particles. If these fine biochar particles have the same warming potential as black carbon, we estimated using black carbon's GWP20 characterization factor that emissions 0.3–6.7 kg ha 1 (0.01–0.26% of 2.5 tonnes ha^{-1} application rate) of biochar dust can result in no net negative emissions for the biochar.

These fine particulate matter emissions from biochar are also relevant for air quality and associated health risks. Proper assessment of the amount of fine biochar particles emitted during its application and from the field are still to be performed. But mitigation of those emissions exists such as applying wet biochar or as slurry and quick incorporation in the soil layer, cover crop canopy and limited tillage of the fields will also limit emissions of such particles with benefit regarding climate and air quality.

5.7 Conclusion

Pyrolysis of biomass is a well-known process capable of producing valuable product for agriculture and energy production. It can use low-value by-products of agricultural and forestry activities, leading to low land requirements. Contrary to implementing practices aiming at increasing soil carbon stock on cropland that need to be sustained over time, biochar does not represent an additional work for the farmer besides its initial incorporation into the soil. Biochar can provide agronomic co-benefits across most of Europe in the 30 next years, in particular as moderate application rate allow to treat more land therefore providing larger co-benefits, and reducing several impacts from the application of nitrogen fertilizer, but at the cost of also increasing some other more largely.

Deep and fast cut in GHG emissions are required to achieve the Paris agreement, the sooner net zero emissions are achieved the less reliant will we be on NETs. Biochar can help offset emissions at low cost using currently available feedstock to ramp up NE before new technologies like bioenergy with carbon capture and storage (BECCS) or direct air carbon capture and storage (DACCS) becomes economically viable. However given the scale of climate mitigation that needs to be achieved, the amount of investment that is required to deploy those technologies, the limited amount of non-used biomass and of land to increase production and the level of competition for these resources it is unlikely that NETs will be the climate crisis' solution alone. NETs can be an options to support decarbonization of the economy and offset diffuse emissions, but should not be an excuse to delay or stop effort in increasing energy efficiency, reducing energy losses, changes in diets and other important aspects of effective climate mitigation.

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Chapter 6. Appendices

6.1 Supplementary information to Chapter 2

Supplementary Information

Potentials, limitations, co-benefits and trade-offs of biochar applications to soils for climate change mitigation

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Table S1: Main controlling factors on the effect of biochar on agricultural yield.

Controlling factors		Observations	Ref
Biochar	Feedstock	Sludge and manure biochars have the highest positive crop yield response, followed by herbaceous and last by wood feedstock. In temperate region, all types of feedstock lead to no significant positive effect on yield. Tropical soils: Sludge and manure biochars improve yield on average by 70%. Wood and herbaceous biochars only lead to 20% increase.	(Jeffery et al. 2017; Biederman and Harpole 2013)
	CEC	Higher biochar CEC lead to higher increase in yield.	(Jeffery et al. 2017)
	Pyrolysis	There is no clear effect of pyrolysis temperature on yield response (beside for >650°C that significantly has negative effect on yield). However fast pyrolysis and hydrothermal carbonization lead to significant negative crop yield effect.	(Jeffery et al. 2017)
	Ash content	Ash content in biochar higher than 10% reduces yield increase from 30% to less than 10%.	(Jeffery et al. 2017)
	C/N ratio	Biochar's C/N ratio above 100 (biochars poor in N) lead to overall negative yield response	(Jeffery et al. 2017)
Soil	pH	Tropics: Yield response to biochar application increases as soils are more and more acidic. Temperate: Significant negative yield response (down to -30%) are observed for neutral and alkaline soils.	(Jeffery et al. 2017, Wang et al. 2019)
Management	Application rate	Tropics: Yield response seem to cross a threshold at application rate between 50-150 t biochar per ha. Temperate: Application rate should be kept between 10-50 t biochar per ha, particularly less than 50 t biochar per ha as above that threshold significantly negative yield response is significant.	(Jeffery et al. 2017)
	Fertilizer	Co-application of fertilizer with biochar can increase yield response.	(Jeffery et al. 2017)
CEC: cation exchange capacity; C/N: carbon to nitrogen ratio			

Table S2: Main controlling factors on the effect of biochar on soil and exposition to toxic compounds.

Controlling factors		Observations	Ref
Biochar	Feedstock	Biochar feedstock is the main determinant for heavy metal concentration in biochar; out of 14 heavy metals, manures-biochar had higher concentrations for 11 of them. PAHs concentrations are higher in plant-based biochar, due to higher carbon content than in manures. No clear pattern between feedstocks and VOCs production have been observed. Feedstock rich in chlorine, such as food waste, can lead to production of chlorinated compounds such as dioxins.	(Qiu et al. 2015; Dutta et al. 2017; Hale et al. 2012)
	Temperature	Production: Temperature is the main control for PAHs concentration on biochar, biochars produced at temperrature between 350 and 550°C show higher PAHs concentration than those produced at both lower or higher temperature. Dioxins production is maximum at low temperature (200-400°C). Amount of VOCs adsorbed on biochar surface decreases with increasing temperature, and is really reduced past 500°C. Once in soils: Higher temperature biochars have higher sorption capacity toward pesticide and other organic compounds, mostly due to higher porosity. Liming effect of higher temperature biochars may also be important for reducing toxicity of heavy metals.	(Hale et al. 2012; Dutta et al. 2017; Qiu et al. 2015; Ghidotti, Fabbri, and Hornung 2017; Yavari, Malakahmad, and Sapari 2015)
	Reaction time	Reaction time is an important control for the production of toxic compounds during pyrolysis. Short reaction time (e.g. fast pyrolysis) produces biochars that contain higher concentration PAHs, VOCs, and dioxins at their surface.	(Dutta et al. 2017; Hale et al. 2012)
Soil	Soil carbon	Soil carbon is an important control for the sorbing capacity of soils. Soils with high carbon content are likely to be able to sorb pollutants desorbed by biochars.	(Dutta et al. 2017)
	Pollution status	Prior level of contaminant in soils may already saturate its sorbing capacity, allowing higher desorption of biochars' contaminant.	(Dutta et al. 2017)
PAH: Polycyclic aromatic hydrocarbons; VOC: volatile organic carbon			

Table S3: CDR requirements for different temperature pathways. Taken from Huppmann et al. (2018)

Carbon sequestration (GtCO2/year)

Deploy. horizon	**2030**						**2050**						**2100**				
Temp. pathway	**Below 1.5C**	**1.5C low overshoot**	**1.5C high overshoot**	**Lower 2C**	**Above 2C**	**Higher 2C**	**Below 1.5C**	**1.5C low overshoot**	**1.5C high overshoot**	**Lower 2C**	**Above 2C**	**Higher 2C**	**Below 1.5C**	**1.5C low overshoot**	**1.5C high overshoot**	**Lower 2C**	**Above 2C**
count	9	43	37	67	167	57	9	44	37	74	175	58	9	44	37	74	180
mean	2.15	2.66	2.03	1.45	0.94	1.21	7.42	11.99	14.92	8.89	3.68	9.24	11.24	18.08	21.84	15.14	11.69
std	2.40	1.85	1.55	1.29	1.22	1.51	5.40	5.29	4.75	5.68	4.12	5.85	7.67	11.02	9.88	10.91	11.78
min	0.13	0.12	0.07	0.00	0.00	0.00	0.31	0.31	5.24	0.00	0.00	0.00	0.07	0.07	2.74	0.00	0.00
5%	0.13	0.38	0.08	0.00	0.00	0.00	1.08	1.69	8.27	0.00	0.00	0.00	0.46	1.77	9.53	0.00	0.00
10%	0.13	0.39	0.49	0.00	0.00	0.02	1.86	5.12	9.34	0.00	0.00	1.66	0.85	6.08	13.45	0.00	0.00
15%	0.19	0.42	0.58	0.00	0.00	0.09	2.74	6.40	9.77	0.56	0.00	3.85	1.99	8.87	14.52	0.43	0.00
25%	0.40	0.80	0.80	0.36	0.00	0.20	4.74	7.61	12.40	6.06	0.01	5.50	5.76	11.95	15.96	8.13	0.23
35%	0.41	1.49	1.12	0.49	0.07	0.46	5.41	10.34	12.83	7.30	1.04	7.05	10.75	14.77	16.29	11.26	3.46
50%	0.42	2.66	1.65	1.09	0.36	0.91	6.25	13.55	15.09	9.58	2.48	9.49	12.96	17.05	22.04	14.91	8.17
75%	3.26	4.14	2.93	2.49	1.46	1.54	9.13	16.02	17.46	12.08	5.71	11.45	14.88	21.68	25.52	21.31	18.40
95%	5.83	5.39	4.38	3.75	3.49	3.21	15.99	18.07	22.41	18.64	11.91	20.13	21.22	30.17	43.57	31.10	35.94
max	5.93	6.30	7.63	4.56	5.77	9.78	17.34	20.21	28.32	24.63	22.90	29.96	23.13	62.87	45.43	45.65	48.73

Table S4: Total cumulative CDR requirements for different temperature pathways. Taken from Huppmann et al. (2018)

Carbon sequestration (GtCO2)

Temperature pathway	Below 1.5C	1.5C low overshoot	1.5C high overshoot	Lower 2C	Above 2C	Higher 2C
count	7	43	35	63	158	51
mean	611.80	828.28	1,042.61	855.82	484.52	983.39
std	196.00	363.49	312.34	351.99	477.39	390.22
min	268.06	0.00	480.78	112.88	0.00	247.13
5%	335.11	126.90	632.32	410.27	0.00	503.11
15%	469.21	427.87	809.53	517.67	0.00	669.45
25%	525.16	558.79	863.92	657.12	3.70	744.97
50%	619.62	903.80	919.60	827.42	413.91	890.97
75%	753.22	1,033.10	1,207.02	1,002.16	819.15	1,174.84
95%	825.71	1,399.39	1,592.01	1,482.78	1,323.81	1,768.92
max	838.19	1,493.92	1,858.40	1,978.85	2,034.77	2190.55

Table S5: Key biochar properties and their controlling factors under slow pyrolysis.

Property	Controlling factor	Observations	Ref
Yield and carbon content	Feedstock	For similar pyrolysis temperature, ligno-cellulosic materials (e.g. woody and herbaceous feedstocks) have higher yield and carbon content, than manures or sludge.	(Li et al. 2019)
	Temperature	Yield decreases with increasing temperature and reaches a plateau at about 600°C for wood/herbaceous feedstock, and 400°C for manures/biosolids. Carbon content in biochar increases linearly with pyrolysis temperatures.	(Li et al. 2019)
	Additives	Potassium (K) increases the amount of carbon retention in biochar by 45%.	(Mašek et al. 2019)
Porosity and surface area	Feedstock	Macroporosity retains the cell structure of feedstock. Woody feedstock have much higher surface area than other feedstocks: Woody > herbaceous > manures > sludge (surface area is divided by a factor two between each categories).	(Li et al. 2019; Wildman and Derbyshire 1991; Gray et al. 2014)
	Temperature	Higher pyrolysis temperature increases biochar's microporosity. Surface area increases with pyrolysis temperature.	(Li et al. 2019)
Ash	Feedstock	Ash content increases, as sludge/digestate > manures > herbaceous > wood. Ash content also changes with feedstock. Herbaceous-derived biochars have higher N and P content than wood-derived biochar, but usually less base cation (Ca^{2+}, Mg^{2+}, K^{+}) (especially compared to softwood). Hardwood-biochar have the most Sulfur. Manures-biochar are both the richest in N and P and base cations.	(Li et al. 2019; Ippolito et al. 2015)
	Temperature	Ash content increases with pyrolysis temperatures.	(Li et al. 2019)
H/C_{org}	Feedstock	Wood and herbaceous feedstock have similar H/C_{org} ratios at a given temperature. Manures and sludge have much higher ratios. Indicate a higher level of aromatic condensation in biochar produced from lignocellulosic biomass.	(Li et al. 2019; Xiao, Chen, and Chen 2016)
	Temperature	H/C_{org} decreases with increasing pyrolysis temperature. Indicate a higher level of aromatic condensation of biochar at higher temperature.	(Li et al. 2019; Weber and Quicker 2018)
O/C_{org}	Feedstock	Wood and herbaceous feedstock have similar O/C_{org} ratios at a given temperature. Manures have much higher ratios.	(Li et al. 2019)
	Temperature	O/C_{org} decreases with increasing pyrolysis temperature	(Li et al. 2019)
C/N	Feedstock	Wood has much higher C/N ratio (much poorer in N compared to C), about twice as high as for herbaceous feedstock. Manures and sludge are much richer in N compared to C, by a factor 10 compared to wood.	(Li et al. 2019)
	Temperature	C/N increases with increasing pyrolysis temperature, biochar becomes poorer in N compared to C.	(Li et al. 2019)
CEC (negative charges on	Feedstock	Wood and manures derived biochars have lower CEC than herbaceous and sludge.	(Li et al. 2019)

biochar)	Temperature	CEC decreases with pyrolysis temperature. Due to lower O/C_{org} and lower H/C_{org}.	(Li et al. 2019)
	pH conditions	Different oxygen functional groups have diffeent pKa, and are deprotonated under different pH conditions. CEC increases with increasing pH, as at higher pH, acids and alcohol of higher pKa are successively deprotonated.	(Banik et al. 2018; Chen et al. 2015; Szymański et al. 2002)
	Aging	During aging in soils, biochar surface is oxidized, increasing O/C_{org} ratios and its CEC.	(Mia, Dijkstra, and Singh 2017)
AEC (positive charges on biochar)	Feedstock	Wood biochar have higher AEC than herbaceouss feedstock.	(Lawrinenko and Laird 2015; Banik et al. 2018)
	Temperature	Pyrolysis temperature increases AEC.	(Lawrinenko and Laird 2015; Banik et al. 2018)
	pH conditions	AEC can be significant in acidic pH but not at neutral or alkaline pH, and quickly decreases with increasing pH. AEC comes mostly from pH dependant sites that are protonated in acidic conditions.	(Lawrinenko and Laird 2015)
	Aging	Very little AEC is structurally stable, and will disappear quickly under soil aging.	(Lawrinenko et al. 2016)
	Additives	Pre-treatment of feedstock with aluminum and iron can increase AEC at higher pH (alkaline conditions).	(Lawrinenko et al. 2017; Banik et al. 2018)
Surface charge	Temperature	The pH at which global charge of surface biochar is null increases with increasing pyrolysis temperature.	(Banik et al. 2018)
pH and alkalinity	Feedstock	Wood biochar are more acidic (range of pH 4-8) than herbaceous (range of pH 6-12). Manures are the most alkaline (range of pH 8-10). Wood biochar have little alkalinity, while herbaceous and manures have higher level of alkalinity. Carbonates in ashes are a major source of alkalinity. Structural low-pKa acid groups ($5 < pKa < 6.4$) can be an important source of alkalinity at low temperature for herbaceous feedstock Wood-derived biochars have higher base cation concentration than herbaceous-derived biochars; but much lower than manures.	(Li et al. 2019; Fidel et al. 2017; Ippolito et al. 2015)
	Temperature	Temperature increases biochars pH and alkalinity. Structural alkalinity decreases with increasing temperature, while temperature increases the amount of carbonates produced during pyrolysis.	(Li et al. 2019; Fidel et al. 2017)
Conductivity	Feedstock	Higher mineral content in herbaceous biochar was linked to a higher electron exchange capacity	(Klüpfel et al. 2014)
	Temperature	Wood charcoal was shown to act as an insulator, a semiconductor and a conductor, respectively at <300 °C, 300 °C–800 °C and >800 °C.	(Joseph et al. 2015; Klüpfel et al. 2014)

		Redox activity is controlled by electronic donating phenolic moeities for low temperature biochars, electron accepting quinone moieties for mid-temperature, and quinone and possibly aromatics for high-temperature biochars.	
C_{org}: organic carbon (exclude carbon present in biochar's ash as carbonates); CEC: cation exchange capacity; AEC: anion exchange capacity			

Table S6: Main controlling factors of the effect of biochar on SOC priming.

Controlling factors		Observations	Ref
Biochar	Feedstock	Crop and wood derived biochar significantly induce negative priming. Sludges and manures derived biochars lead to positive priming mostly. Grass biochars lead to both positive and negative priming with null net effect overall.	(Ding et al. 2018)
	Temperature	Pyrolysis temperatures above 500°C lead to significant negative priming. Negative priming is much stronger for pyrolysis temperature above 600°C.	(Ding et al. 2018)
	Nitrogen	Increasing C/N ratio of biochar increases negative priming. Biochar with more than 4% nitrogen switch from negative to positive priming, however variations in response are important and not statistically significant.	(Ding et al. 2018)
	Carbon	Pyrolysis time, temperature and feedstock control the amount of carbon in biochars. A carbon content over 50% in biochar lead to significant negative priming.	(Ding et al. 2018)
	Aging	Charcoal deposits in historical stabilizes recent input of carbon better than adjacent soils without charred materials.	(Kerré et al. 2016; Hernandez-Soriano et al. 2016; Kerré, Willaert, and Smolders 2017)
Soil	SOC	Soils with less than 1% SOC lead to significant positive priming.	(Ding et al. 2018)
	C/N ratio	Negative priming is larger at soil C/N below 11-12. Above that value, negative priming is not statistically significant.	(Ding et al. 2018)
	Texture	Soil texture seems to have little effect on priming of SOC, being overall negative. Negative priming is more important at clay content above 50%.	(Ding et al. 2018; J. Wang, Xiong, and Kuzyakov 2016)
	pH	Negative priming is more important at soil pH above 6 and slightly decreases with increasing soil pH.	(Ding et al. 2018)
SOC: soil organic carbon; C/N: carbon to nitrogen ratio			

Table S7: Main controlling factors of the effect of biochar on soil methane (CH_4) emissions or uptake.

Controlling factors		Observations	Ref
Biochar	Feedstock	Different meta-analysis draw different conclusion regarding the effect of biochar's feedstock on soil methane emissions.	(He et al. 2017; Ji et al. 2018; Jeffery et al. 2016)
	Temperature	Increasing pyrolysis temperature decreases methane release from 'methane source' soils, but also the oxidative potential of 'methane sink' soils.	(Ji et al. 2018)
	pH	Upland soils see reduced methane uptake for biochars with pH below 7 to a positive increase in uptake for biochars with pH >9.	(Ji et al. 2018; He et al. 2017)
Soil	Moisture	Flooded soils (paddy rice) see their methane emissions reduced after biochar amendment. Upland soils that are 'methane source' see their emissions reduced. Upland soils that are 'methane sink' see their sink capacity reduced.	(Ji et al. 2018; Jeffery et al. 2016)
	pH	In upland soils, biochar reduced methane uptake in acidic and neutral soils. In flooded soils, biochar increases methane release in acid soils, but decreases it in neutral and alkaline soils.	(Ji et al. 2018; Jeffery et al. 2016)
	Texture	Biochar has more pronounced effect on medium textured soils: reducing soil methane emissions from 'methane source' soils and reducing methane sink capacity from 'methane sink' soils. Fine soils see their methane emissions increase after biochar treatment.	(Ji et al. 2018)
Management	Fertilization	Unfertilized soils show higher response to biochar addition regarding decreasing release and uptake of methane. Upon application of organic-N fertilizer, biochar increases the sink capacity 'methane sinks' Both application of organic or synthetic N, decrease emissions of 'methane source' after application of biochar. N-fertilization rate may also influence soil response to biochar application: below 120kgN/ha increasing methane sink/decreasing methane emissions, while more than 120 kgN/ha has opposite effect.	(Jeffery et al. 2016; Ji et al. 2018)
	Application rate	Biochar application rate below 20 t/ha may be beneficial for enhancing soil methane sinks, but increase methane emissions from 'methane source' soils. After that threshold, increasing biochar application rate decreases both methane emissions from source, and decreases sink potential of sinks.	(Ji et al. 2018)

Table S8: Main controlling factors on the effect of biochar on soil nitrogen availability.

Controlling factors		Observations	Ref
Biochar	Feedstock	Crop residues are feedstocks that most significantly reduce availability of nitrogen in agroecosystems.	(Gao, DeLuca, and Cleveland 2019)
	Temperature	Immobilization of both NO_3^- and NH_4^+ is minimal at medium pyrolysis temperature (400-600°C)	(Gao, DeLuca, and Cleveland 2019; T. T. N. Nguyen et al. 2017)
	AEC and CEC	NO_3^- preferentially sorb on AEC sites, while NH_4^+ on CEC sites.	(Ippolito et al. 2015)
	Aging	Via decrease in AEC and increase in CEC.	(Ippolito et al. 2015)
Soil	pH	Soil pH is noted as being an important controlling factor of biochar's effect on soil nitrogen availibility by both Gao and colleagues and Nguyen and colleagues, but they disagree.	(Gao, DeLuca, and Cleveland 2019; T. T. N. Nguyen et al. 2017)
Management	Fertilizer	Biochar reduces NH_4^+ availability under all N-fertilizer type, but organic-N application. Co-application of fertilizer and biochar may reduces risk of immobilizing nitrate after biochar application. Urea and NH_3-based fertilizers have potential to mitigate nitrate deficiency.	(Gao, DeLuca, and Cleveland 2019; T. T. N. Nguyen et al. 2017)
	Application rate	Increasing application rate of biochar tend to reduce nitrogen availability.	(Gao, DeLuca, and Cleveland 2019; T. T. N. Nguyen et al. 2017; Borchard et al. 2019)

Table S9: Main controlling factors of the effect of biochar on soil ammonia (NH_3) volatilization rate.

Controlling factors		Observations	Ref
Biochar	pH	Biochars with pH above 9 statistically increase NH_3 volatilization. Biochar pH below 7 tend to increase NH_3 volatilization as well. In between it may or may not decrease NH_3 volatilization.	(Liu et al. 2018)
	Feedstock	Manure biochars increase NH_3 volatilization, because of their high alkalinity. Biochar from woody biomass reduces NH_3 volatilization more efficiently because of higher sorption capacity.	(Liu et al. 2018; Sha et al. 2019)
	Aging	Aging of biochar in soils increases its CEC and Biochar liming effect is only temporary. Enhancement of ammonia volatilization after biochar application is expected to be only temporary, in the long term biochar may reduce soil NH_3 volatilization.	in text begining p.219 (Liu et al. 2018)
Soil	pH	Biochar particularly increases NH_3 volatilization in acidic soils with pH below 5-6 and has little effect, even potentially reducing emissions over the rest of the pH range.	(Liu et al. 2018; Sha et al. 2019)
	Native SOC	Biochar applied to soils with low native organic carbon (<2%) increase NH_3 volatilization Above 3% SOC, biochar reduction in NH_3 volatilization is statistically significant.	(Liu et al. 2018; Sha et al. 2019)
	Texture	The 2 meta-analysis available contradict each other regarding the effect of biochar in fine soil: Sha et al. (2019) find a significant decrease in NH_3 volatilization in finer soils, while Liu et al. (2018) a significant increase. Liu et al.'s explanation of reduced resistance to volatilization due to higher aeration in fine soil after biochar application is a compelling argument in their case.	(Liu et al. 2018; Sha et al. 2019)
	CEC	Soils with low CEC see enhancement volatilization upon biochar application. This enhancement decreases with increase soil CEC.	(Liu et al. 2019)
Management	Application rate	Higher application rate tend to increase NH_3 volatilization compared to controls, usually explained by a higher liming effect.	(Liu et al. 2018; Sha et al. 2019)
	Fertilizer	Biochar application may be able to handle low N-fertilization (<200kg/ha) as an overall decrease in ammonia volatilization is observed, but higher fertilizer application rate will see higher NH_3 volatilization. Using organic fertilizer or urea do not enhance NH_3 volatilization, but ammonium fertilizer does.	(Sha et al. 2019)

Table S10: Main controlling factors on the effect of biochar on soil phosphorus availability.

Controlling factors		Observations	Ref
Biochar	Feedstock	Biochar derived from crop residues and manures increase the most phosphorus availability.	(Gao, DeLuca, and Cleveland 2019)
	Temperature	Biochar produced at lower pyrolysis temperatures allows for more available phosphorus. At high temperature stable compounds of phosphorus are produced from the feedstock, limiting its supply to plants. Temperature above 600°C may reduce soil P-availability.	(Gao, DeLuca, and Cleveland 2019; Glaser and Lehr 2019)
	Ash	Ca^{2+} and Mg^{2+} present in the biochar ashes can lower availability of phosphorus by precipitation.	(Ippolito et al. 2015)
Soil	Texture	Medium textured soils have higher responses to biochar application, with an increase in phosphorus availability.	(Gao, DeLuca, and Cleveland 2019)
	pH	Soil with pH over 7.5 may experience lower P availability after biochar application, potentially due to liming effect reducing P-availability, or Ca-P precipitation.	(Gao, DeLuca, and Cleveland 2019; Glaser and Lehr 2019)
Management	Application rate	Increasing application rate was found to increase phosphorus availability, mostly as more biochar brings more phosphorus to the soil. Increased availability is significant at application rate above 10 tonnes biochar per hectare. However, at higher application rate, liming (soil pH increase) may be more important.	(Gao, DeLuca, and Cleveland 2019; Glaser and Lehr 2019)

Table S11: Main controlling factors on the effect of biochar on soil nutrient leaching.

Controlling factors		Observations	Ref
Biochar	Ash	Nutrient brought with biochar can readily be leached out of soils.	(Ippolito et al. 2015)
Soil	Hydraulic conductivity	Increase in soil hydraulic conductivity after biochar application can lead to an increase in the amount of leached nutrients, if they are made more available after biochar application. As such, biochar may reduce nutrient leaching in coarser soil, and may increase it in finer soils.	(Laird and Rogovska 2015)
Management	Application rate	In case of leaching of biochar's nutrient, higher application rate will lead to more leaching. Application rate will also modulate leaching rate as they affect nutrient availability and soil water retention and conductivity.	(Ippolito et al. 2015; Laird and Rogovska 2015)

Table 12: Main controlling factors on the effect of biochar on soil water availability and soil hydraulic conductivity.

Controlling factors		Observations	Ref
Biochar	Hydro-phobicity	Biochar hydrophobicity is an important control on its water uptake potential.	(Gray et al. 2014; Kinney et al. 2012)
	Porosity	Biochar's macroporosity is more important for water retention than its microporosity. Choice of feedstock with appropriate macrostructure is important.	(Gray et al. 2014; Kinney et al. 2012)
	Particle size and shape	Depending on biochar particle size, biochar can clog or increase the size of soil pores. Clogging happen when biochar particle size is lower than the size of soil interpores. As a consequence, both soil hydraulic conductivity and soil water availability increases. Shape of biochar particles has been suggested to influence soil's interpores structure by disrupting and modifying the interpores size distribution. But very fine biochar particles can loose their porosity, in particular macroporosity.	(Sun and Lu 2014; Trifunovic et al. 2018)
Soil	Water repellency	Biochar's effect on soil water repellency has been little studied. Most of the studies reported no effect of biochar, a few reported conflicting results.	(Blanco-Canqui 2017; Hallin et al. 2015)
	Texture	Coarse soil see higher increase in soil water availability than finer soil, due to increase in mesopore that allow retention of water after biochar application. Biochar reduces water infiltration and hydraulic conductivity in coarse soil, and the opposite for fine soils. Thus improving soil hydrology in both cases, though the effect is usually more important in coarse soils. Medium textured soils receive less benefit from biochar application.	(Omondi et al. 2016; Blanco-Canqui 2017)
	Soil pore size	Soil pore size distribution is important for soil hydrology. Biochar may influence soil pore size distribution via its particles size and shape, and its effect on soil aggregates. Biochar has effect on aggregate stability, which may prevent clogging, and aggregate size, and may increase soil pore size.	(Sun and Lu 2014; Trifunovic et al. 2018; Blanco-Canqui 2017)
	Run-off volume	Biochar reduced run-off volume in 4 out of 6 studies, from 5 to 50% reduction.	(Blanco-Canqui 2018)
Manage-ment	Application rate	Soil available water capacity increases with increasing biochar application rate. But a minimum application rate may be required to observe significant response. Most studies that observed increase in soil available water used application rate >25 t biochar/ha. Soil saturated hydraulic conductivity increases with biochar application rate, but a minimum amount of biochar may be required before significant response (~>20 t biochar/ha).	(Omondi et al. 2016; Blanco-Canqui 2017)

Table S13: Classification of the LCA studies under type of feedstock and origin (residues, dedicated plantations or waste) and for which life-cycle stages the results were used in figure 3 in the main text.

References	**Herbaceous**	**Wood**	**Organic waste**		**Residue**	**Dedicated plantation**	**Waste**		**Supply-chain**	**Avoided emissions**	**Carbon sequestration**	**Effects on soils**	**Total**
(Roberts et al. 2010)	**x**	**x**			**x**	**x**	**x**		**x**	**x**	**x**	**x**	**x**
(Hammond et al. 2011)	**x**	**x**			**x**	**x**			**x**	**x**	**x**	**x**	**x**
(Ibarrola, Shackley, and Hammond 2012)	**x**	**x**	**x**		**x**		**x**		**x**	**x**	**x**	**x**	**x**
(Meyer et al. 2012)	**x**	**x**			**x**				**x**	**x**	**x**	**x**	**x**
(Field et al. 2013)		**x**			**x**				**x**	**x**	**x**	**x**	**x**
(Lugato et al. 2013)		**x**			**x**								
(T. L. T. Nguyen, Hermansen, and Nielsen 2013)	**x**				**x**								
(Cao and Pawlowski 2013)			**x**				**x**						**x**
(Sparrevik et al. 2013)	**x**				**x**								
(Z. Wang et al. 2014)	**x**				**x**				**x**	**x**	**x**	**x**	**x**
(Sparrevik et al. 2014)		**x**			**x**								
(Peters, Iribarren, and Dufour 2015)		**x**				**x**							**x**
(Homagain et al. 2015)		**x**			**x**								**x**
(Thornley et al. 2015)		**x**				**x**							**x**
(Miller-Robbie et al. 2015)			**x**				**x**						**x**

(Clare et al. 2015)	**x**				**x**								**x**
(Mohammadi et al. 2016)	**x**				**x**								
(Pietro Bartocci et al. 2016)		**x**				**x**							**x**
(Muñoz et al. 2017)	**x**	**x**			**x**								**x**
(Ericsson et al. 2017)		**x**				**x**							
(Smebye et al. 2017)		**x**			**x**								
(Llorach-Massana et al. 2017)	**x**				**x**								**x**
(Robb and Dargusch 2018)	**x**				**x**								**x**
(Mohammadi et al. 2019a)			**x**				**x**						**x**
(Mohammadi et al. 2019b)			**x**				**x**						**x**
(Rajabi Hamedani et al. 2019)		**x**	**x**			**x**	**x**						**x**
(Barry et al. 2019)			**x**				**x**						
(Azzi, Karltun, and Sundberg 2019)		**x**			**x**				**x**	**x**	**x**	**x**	**x**
(Lu and El Hanandeh 2019)		**x**			**x**								**x**
(Tadele et al. 2019)	**x**					**x**			**x**				
(Thers et al. 2019)	**x**					**x**							
(Uusitalo and Leino 2019)	**x**	**x**			**x**								
(Xu et al. 2019)	**x**				**x**								

Table S14: Classification of LCA studies according to the type of impact/indicator they include in the analysis: global warming potential (GWP), acidification potential (AP), ozone depletion potential (ODP), eutrophication (EP), smog formation, respiratory effects (REP), carcinogenic potential (CP), non-carcinogenic potential (NCP), ecotoxicity (ECT), and fossil fuel depletion (FFD), end-points (e.g. Human health, Ecosystem health, Resource depletion), Energy use (e.g. life-cycle energy use, cumulative energy use), cost (e.g. life-cycle cost, environmental valuation)

References	**GWP**	**AP**	**ODP**	**EP**	**REP**	**CP**	**NCP**	**ECT**	**FFD**		**End Points**	**Energy use**	**Cost**
(Roberts et al. 2010)	x											x	x
(Hammond et al. 2011)	x												
(Ibarrola, Shackley, and Hammond 2012)	x												
(Meyer et al. 2012)	x												
(Field et al. 2013)	x												x
(Lugato et al. 2013)	x												
(T. L. T. Nguyen, Hermansen, and Nielsen 2013)	x	x		x	x				x				
(Cao and Pawlowski 2013)	x											x	
(Sparrevik et al. 2013)	x				x				x				
(Z. Wang et al. 2014)	x												
(Sparrevik et al. 2014)	x				x								
(Peters, Iribarren, and Dufour 2015)	x	x		x								x	
(Homagain et al. 2015)		x			x								
(Thornley et al. 2015)	x	x	x	x	x	x	x	x	x		x		
(Miller-Robbie et al. 2015)			x				x					x	
(Clare et al. 2015)	x				x								x
(Mohammadi et al. 2016)	x												
(Pietro Bartocci et al. 2016)	x												
(Muñoz et al. 2017)	x			x	x	x	x		x				
(Ericsson et al. 2017)	x											x	

(Smebye et al. 2017)											x		
(Llorach-Massana et al. 2017)	x												
(Robb and Dargusch 2018)	x												
(Mohammadi et al. 2019a)	x	x	x	x	x	x	x	x	x				
(Mohammadi et al. 2019b)	x	x	x	x	x	x	x	x	x				
(Rajabi Hamedani et al. 2019)	x	x	x	x	x	x	x	x	x		x	x	x
(Barry et al. 2019)	x							x					
(Azzi, Karltun, and Sundberg 2019)	x												
(Lu and El Hanandeh 2019)	x											x	x
(Tadele et al. 2019)	x	x	x	x	x	x	x	x	x				
(Thers et al. 2019)	x												
(Uusitalo and Leino 2019)	x												
(Xu et al. 2019)	x												

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6.2 Supplementary information to Chapter 3

Supplementary information
Life-cycle assessment to unravel co-benefits and trade-offs of large-scale biochar deployment in Norwegian agriculture

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Table S1: Inventories for barley production in Norway

	Barley cultivation in Norway without biochar, ha.year	*Barley cultivation in Norway with biochar, ha.year*	*Barley cultivation in Norway with biochar-fertilizer, ha.year*	
<u>Output to technosphere</u>				
	1	1	1	ha.year
<u>Inputs from technosphere</u>				
tillage, ploughing	1	1	1	ha.year
sowing	1	1	1	ha.year
tillage, harrowing, by rotary harrow	2	2	2	ha.year
fertilising, by broadcaster	1	1	1	ha.year
application of plant protection product, by field sprayer	2.333	2.333	2.333	ha.year
combine harvesting	1	1	1	ha
rolling	1	1	1	ha
liming	447	302	302	kg
cyclic N compound	0.075	0.075	0.075	kg
default pesticide inventory	0.01	0.01	0.01	kg
triazine compound	0.011	0.011	0.011	kg
pyretroid compound	0.017	0.017	0.017	kg
benzoic compound	0.088	0.088	0.088	kg
organo phosphorus coompound	0.98	0.98	0.98	kg
packaging, for pesticides	1.181	1.181	1.181	kg
Ammonium nitrate, as N	112	112		kg
Ammonium nitrate, as N	15.5	15.5		kg
Phosphate fertiliser, as P2O5	39.5	39.5		kg
Potassium chloride, as K2O	75.9	75.9		kg
packaging, for fertilisers	426.3	426.3		kg
Biochar application (spreading and harrowing)		2552	2552	kg
Biochar		2552		kg
Biochar-fertilizer			2552	kg
barley seed, for sowing	160	160	160	kg
transport, tractor and trailer, agricultural	30	30	30	tkm
<u>Emissions to air</u>				

Ammonia	7.75	8.13	8.13	kg
Dinitrogen monoxide	2.43	1.65	1.65	kg
Nitrogen oxides	5.1	4.59	4.59	kg
Emissions to soil				
Fludioxonil	0.01	0.01	0.01	kg
Tribenuron-methyl	0.011	0.011	0.011	kg
Alpha-cypermethrin	0.017	0.017	0.017	kg
Trifloxystrobin	0.088	0.088	0.088	kg
Prothioconazol	0.075	0.075	0.075	kg
Ethepan	0.05	0.05	0.05	kg
Glyphosate	0.93	0.93	0.93	kg
Cadmium	4.83E-03	4.83E-03	4.83E-03	kg
Chromium	3.79E-03	3.79E-03	3.79E-03	kg
Lead	9.85E-04	9.85E-04	9.85E-04	kg
Nickel	4.14E-03	4.14E-03	4.14E-03	kg
Emissions to water				
Cadmium, ion, groundwater	4.39E-05	4.39E-05	4.39E-05	kg
Cadmium, ion, river	2.81E-05	2.81E-05	2.81E-05	kg
Chromium, ion, groundwater	1.86E-02	1.86E-02	1.86E-02	kg
Chromium, ion, river	2.81E-03	2.81E-03	2.81E-03	kg
Copper, ion, groundwater	2.60E-03	2.60E-03	2.60E-03	kg
Copper, ion, river	1.93E-03	1.93E-03	1.93E-03	kg
Lead, groundwater	1.65E-04	1.65E-04	1.65E-04	kg
Lead, river	3.82E-05	3.82E-05	3.82E-05	kg
Nickel, ion, river	1.61E-03	1.61E-03	1.61E-03	kg
Nitrate	28.7	25.8	25.8	kg
Zinc, ion, groundwater	2.27E-03	2.27E-03	2.27E-03	kg
Zinc, ion, river	1.13E-02	1.13E-02	1.13E-02	kg

Table S2: Proximate, ultimate and sulfate analysis and higher heating values (HHV) of the feedstock, biochar and tar (i.e. organic fraction of the bio-oil).

	Feedstock	Biochar	Tar
Fixed matter (% db)	15.0	80	32.1
Volatile matter (% db)	84.2	17.0	67.9
Ash (% db)	0.8	3.0	0
C (% db)	49.0	79.9	61.4
H (% db)	5.8	2.5	7.3
N (% db)	0.1	0.1	0.2
Cl (% db)	0.01	0.01	0.003
S (% db)	0.02	0.04	0.01
O (% db)	44.2	14.4	31.0
Pyritic (% db)	0	0	0
Sulfate (% db)	0	0	0
Organic (% db)	0.02	0.04	0.01

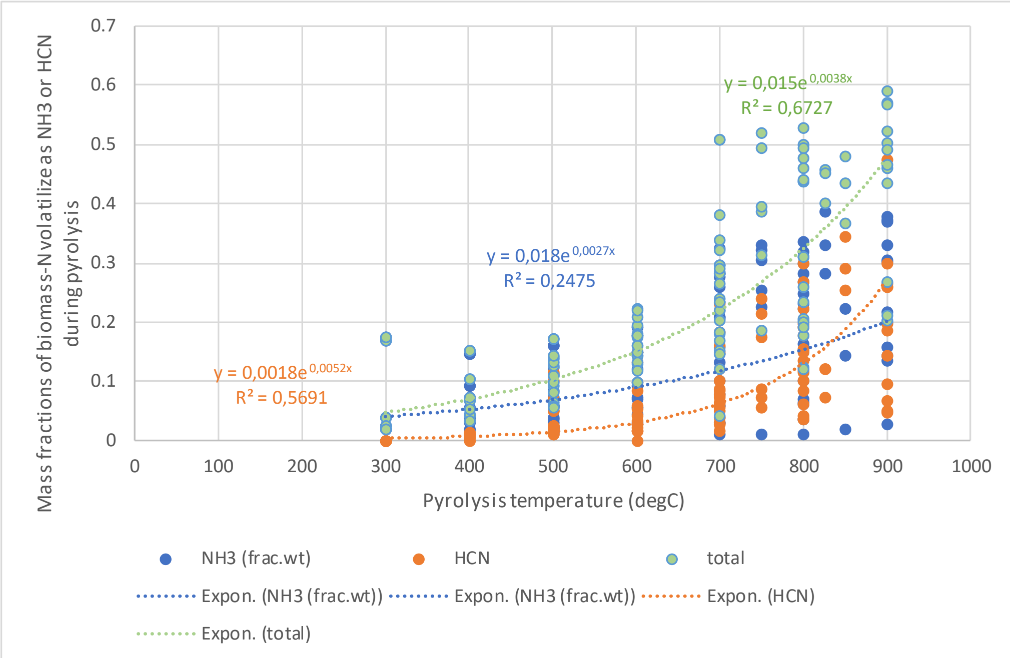

Figure S1: Biomass-N volatilization in NH3 and HCN during pyrolysis. Based on data from (Chen et al. 2012; Zhan et al. 2017; Abelha, Gulyurtlu, and Cabrita 2008; Becidan, Skreiberg, and Hustad 2007; Zhan et al. 2019)

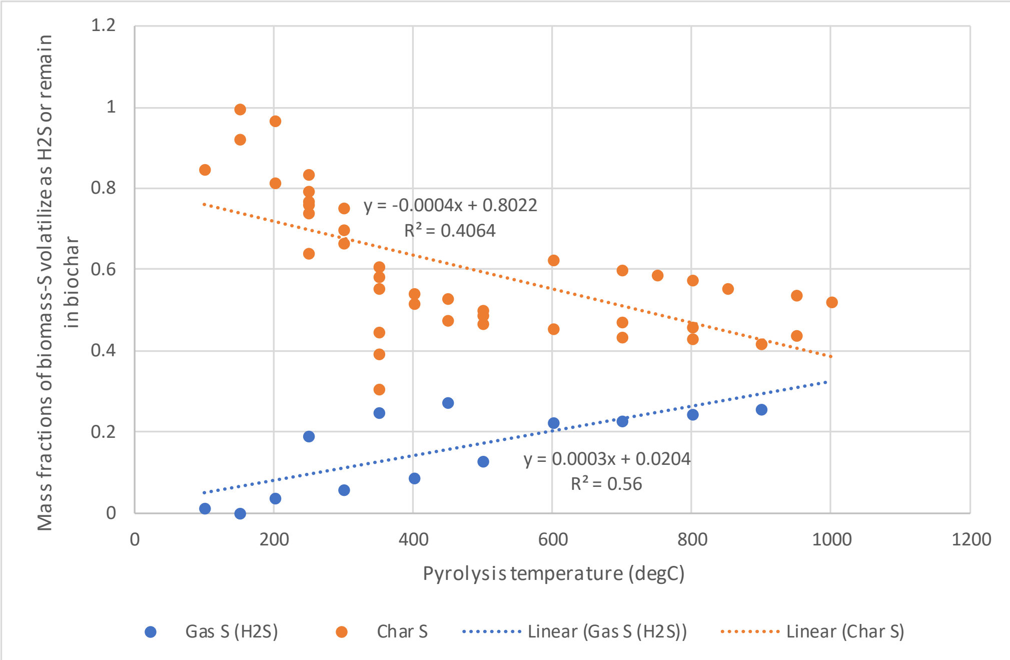

Figure S2: Biomass-S fate between biochar-S and gas-S during pyrolysis. Based on data from (Knudsen et al. 2004; Saleh et al. 2014; Zhang et al. 2017; Liu et al. 2015)

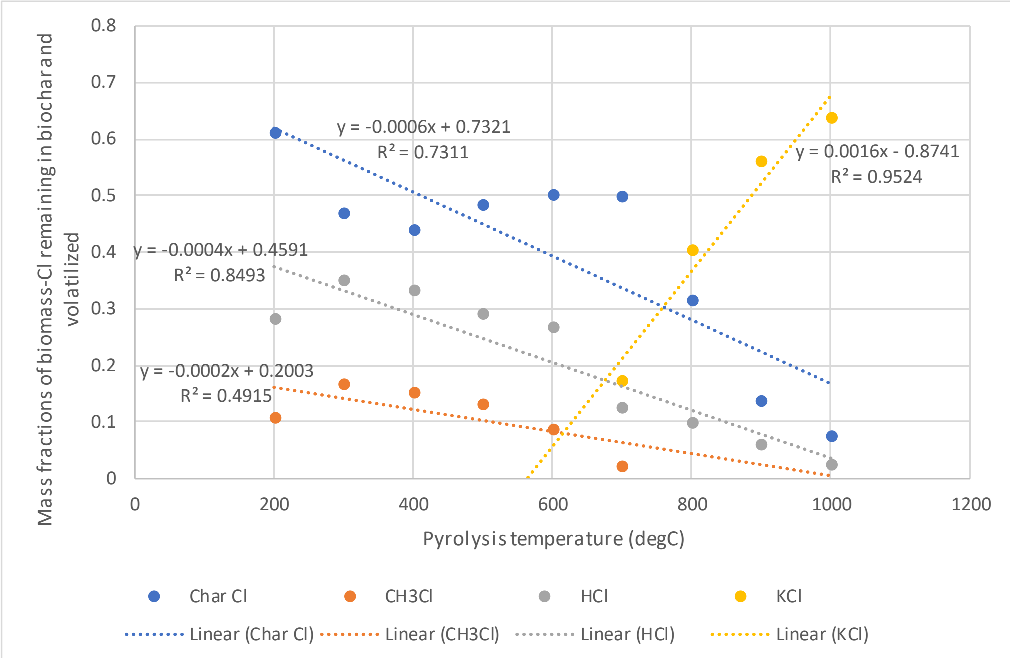

Figure S3: Biomass-Cl fate between biochar-Cl and gas-Cl during pyrolysis. Based on (Peng et al. 2019)

Table S3: Yields of the products of pyrolysis. Yields are given on a dry matter basis.

	Yields (kg/kg biomass db)
Biochar	0.28
Tar	0.34
CO	0.04
H2	3.8E-04
CH4	0.01
C2H2	2.2E-03
CO2	0.11
NH3	1.1E-04
HCN	6.3E-05
H2S	3.2E-05
HCl	2.0E-05
CH3Cl	9.5E-06
Water	0.22

Supplementary text 1

Biomass feedstock, biochar and tar (i.e. organic fraction of the bio-oil) are modeled as non-conventional component. A drawback to modeling tar as a non-conventional component is that such component does not participate in phase equilibrium and does not change phase. As such, it is unable to account for the latent heat of vaporization of the tar.

The enthalpy of the tars at the output of the pyrolysis reactor is underestimated as it does not account for the latent heat of vaporization of the tars that leave the reactor as vapors. For this same reason the duty of the pyrolysis (i.e the energy required to perform the pyrolysis) is also under-estimated. To overcome this limitation, a heat flow is subtracted to the pyrolysis reactor that represent the latent heat of vaporization of the tar, via a calculator block. This estimate for the latent heat of vaporization of tars of 1.22 MJ/kg used in (Woolf et al. 2014, 204). By doing so, we find a pyrolysis duty of about 0.0785 MJ/MJ feedstock, which is in the range of estimated values in the literature 0.06 to 0.15 MJ/MJ feedstock (Crombie and Mašek 2014).

Figure S4 shows the Aspen simulation for the pyrolysis with CHP case:

- DRYER dries the incoming wet biomass at 40% moisture to 10% moisture. DRY-BIO then enters the pyrolysis reactor (PYRO), which is modeled as a RYield reactor, where input of yield and composition of biochar and tar are entered as indicated in Table S2.
- L1 represents the heat of vaporization of the tars. It is accounted as a negative heat flow entering the reactor.
- SEP split the biochar, tar and syngas flows. Biochar is directly recovered. Syngas is sent directly for combustion. Tar is sent to the DECOMP reactor.
- Tar is modeled as a non-conventional compound and procedure for combustion of non-conventional compound is followed (AspenTech 2013). The tars are decomposed in the RYield DECOMP into elemental C, O2, H2, N2, elemental S and Cl2, based on the ultimate analysis. The decomposed tar (DEC-TAR) enters the combustion reactor BURNER via INBURNER flow to be combusted together with the syngas. DEC-DUTY heat flow between DECOMP and BURNER transfer the heat of reaction from the decomposition to the combustion.
  The DECOMP reactor also has an external heat stream (L2) which correspond to the estimated latent heat of vaporization of the tars, and represents the surplus of enthalpy contained in the vaporized tars, which is not accounted for by Aspen.
- Combustion in BURNER is conducted with air (assumed 79% N2 and 21% O2) at a temperature of 1000°C. A 3% excess of oxygen for the combustion is assumed (Sørmo et al. 2020). Larger excess reduces CO emissions but increases NOx emissions, and vise versa. The stoichiometric amount of oxygen required for complete combustion is calculated based on the flow of CO, CH4, C2H2 and H2 from SYNGAS and the flow of C and H2 from INBURNER (decomposed tar). Then the amount of O2 (coming from the decomposition of the tars) from INBURNER is subtracted and the air-excess coefficient is applied.
- The combustion reaction is modeled in a RGibbs reactor, which determines reactions and yields based on the minimization of the free Gibbs energy and therefore assume that the combustion reaction reaches equilibrium. The list of potential products of combustion considered for the RGibbs reactor are H2O, N2, O2, NO2, NO, N2O, S, SO3, SO2 Cl2, HCl C, CO, CO2, H2, CH4, C2H2.
- Energy generated during the combustion of the tars and gas in BURNER and from cooling the flue gas from 1000 to 120°C is recovered after subtracting the energy required by the PYRO duty (i.e. energy required for the pyrolysis).

- The co-generation steam cycle is not modeled in Aspen, rather, standard conversion value from industry is used. A electricity-to-heat ratio of 0,4 is used corresponding to an electricity efficiency of 28,5% and a heat efficiency of 71,5% (Sipilä 2016).

Figure S5 shows the Aspen simulation for the pyrolysis with bio-oil recovery case:

- The biomass feed to the pyrolysis reactor and the combustion conditions are similar to the case of pyrolysis with CHP.
- We find that the energy contained in the syngas does not cover the energy requirement to perform the pyrolysis, which is expected according to (Crombie and Mašek 2014). To be able to run the pyrolysis without relying on external fossil fuel to provide the energy to the pyrolysis reactor, we by-pass part of the tar to be burnt. We estimate that about 11% of the tars are needed to supply heat to the pyrolysis. The by-passed tars follow the same procedure as in the previous case for its combustion.
- The remaining vapors from the pyrolysis (tars and gas) are cooled down in the OILCOND heat exchanger where the tars and water are assumed to be condensed down to a temperature of 15°C, which is a typical temperature used for condensing bio-oils in pilot reactors (Papari and Hawboldt 2018). In case condensing down to 15°C is not enough to recover all the bio-oil, the uncondensed compounds would be sent to the combustion, thus reducing the need for the by-pass. One can see the by-pass as accounting for losses of tars regarding both the energy efficiency of the process and potential losses during the condensation.
- As in the first simulation, the latent heat of vaporization of the tar is accounted as a negative heat flow (L1) in the pyrolysis reactor. L2 represents the latent heat of condensation of the 89% of the tars that are condensed in the condenser, while L3 represents the enthalpy of vaporization contained in the tars that goes to the DECOMP reactor (11% of the tars).
- It is assumed that the energy recovered during the condensation of the bio-oil and the energy recovered from the flue gas by cooling it down to 120°C can be recovered and used to dry the incoming biomass.

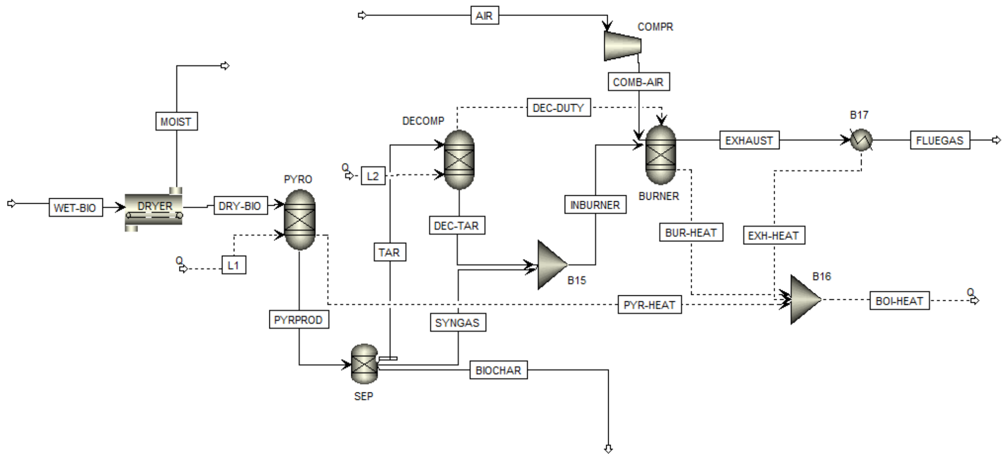

Figure S4: Aspen simulation flowsheet for pyrolysis with combined heat and power (CHP)

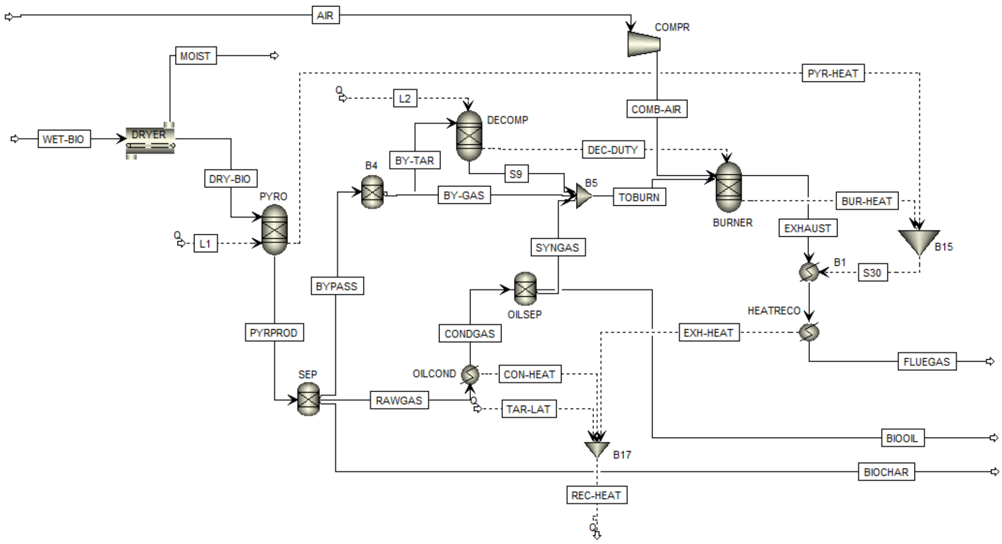

Figure S5: Aspen flowsheet for pyrolysis with bio-oil recovery.

Table S4: Inventories for the different biochar production scenarios

Biochar production scenarios	*with no upgrade of co-products*	*with CHP*	*with sequestration of bio-oil*	unit
Output to technosphere				
Biochar	1	1	1	kg
Electricity		2.1		kWh
Heat		15.9		MJ
Inputs from technosphere				
Wood chips, FR, m3, at regional storehouse	0.0247	0.0247	0.0247	M3
Synthetic gas factory	1.23E-09	1.23E-09	1.23E-09	unit
Electricity	0.281817		0.26	kWh
Bio-oil sequestration			2.1	kg
Transport, freight, lorry >32 metric ton, euro6	2840	2840	2840	tkm
Emissions to air				
CO, biogenic	3.57E-06	3.57E-06	1.95E-06	kg
CO2, biogenic	3.45E+00	3.45E+00	1.03E+00	kg
HCL	1.13E-04	1.13E-04	8.26E-05	kg
H2O	3.94E+00	3.94E+00	2.24E+00	kg
NOx	5.59E-04	5.59E-04	5.93E-05	kg
N2O	4.21E-08	4.21E-08	4.39E-09	kg
SO2	4.87E-04	4.87E-04	2.41E-04	kg
Cl2	3.53E-11	3.53E-11	7.05E-11	kg
H2	2.21E-07	2.21E-07	5.87E-08	kg
PAH, polycyclic aromatic hydrocarbons	0.0398	0.0398	0.004378	mg
As	3.5	3.5	0.175	mg
Cd	0.54	0.54	0.027	mg
Cr	9.6	9.6	0.48	mg
Cu	2	2	0.1	mg
Pb	1.1	1.1	0.055	mg
Hg	0.14	0.14	0.007	mg
Mo	0.57	0.57	0.0285	mg
Ni	1.8	1.8	0.09	mg
Sn	0.03	0.03	0.0015	mg
NMVOC	340	340	17	mg
PM10	1790	1790	89.5	mg
VOC (terpene)	56	56	56	mg

Table S5: Inventory for bio-oil sequestration in geological deposits

	Amount	**Unit**
Output to technosphere		
Sequestration of bio-oil in geological deposits, kg	1	kg
Inputs from technosphere		
Petroleum {NO} petroleum and gas production, off-shore	1	kg
Transport, freight, sea, transoceanic tanker	1.44	tkm
Transport, freight, lorry >32 metric ton, euro6	0.42	tkm

Table S6: Inventory for biochar-fertilizer production

	Amount	Unit
Output to technosphere		
Biochar fertilizer	1	kg
Inputs from technosphere		
Electricity, medium voltage {NO}| market for | APOS, U	0.21	kWh
Ammonium nitrate, as N	4.39E-02	kg
Ammonium nitrate, as N	6.06E-03	kg
Phosphate fertiliser, as P2O5	1.55E-02	kg
Potassium chloride, as K2O	2.47E-02	kg
Emissions to air		
Particulates, >10um	29.25	mg
Particulates, < 2.5um	103.35	mg
Particulates, > 2.5 um, and < 10um	62.4	mg
Lead	9.75E-04	mg
Cadmium	3.90E-05	mg
Copper	4.88E-03	mg
Chromium	2.15E-03	mg
Mercury	1.37E-05	mg
Nickel	9.75E-04	mg
Zinc	1.93E-02	mg

Table S7: Inventories for the application of biochar or biochar-fertilizer at 2552 kg biochar/ha

	Amount		**Unit**
Output to technosphere	**biochar**	**biochar-fertilizer**	
Biochar application (spreading and harrowing)	1	1	kg
Inputs from technosphere			
Fertilising, by broadcaster	3.9E-04	3.9E-04	ha
Tillage, harrowing, by rotary harrow	3.9E-04	3.9E-04	ha
Transport, freight, lorry >32 metric ton, euro6	0.29	0.29	t.km
Emissions to soil, agricultural			
Cadmium	1.62E-05	1.62E-05	kg
Lead	7.02E-05	7.02E-05	kg
Zinc	4.32E-04	4.32E-04	kg

Table S8: Values used for uncertainty analysis regarding soil emissions

		Baseline	Low bound	High bound	Unit	Reference
Direct soil emissions	N2O from fertilizer	0.01	0.008	0.012	kg N2O-N/kg N	+/-20%, (Miljødirektoratet 2019)
	NOx from fertilizer	0.04	0.005	0.104	kg NOx/kg N	(Miljødirektoratet 2019)
	NH3 from fertilizer	0.05	0.04	0.06	kg NH3-N/kg N	+/-20%, (Miljødirektoratet 2019)
	N leaching	0.22	0.19	0.34	kg NO3-N/kg N	Average value (Miljødirektoratet 2019) Min and max values for N-leaching from grain field, calculated from (Bechmann et al. 2017)
Indirect soil emissions	Indirect N2O from N leaching	0.0075	0.00225	0.01275	kg N2O-N/kg N	+/- 70%, (Miljødirektoratet 2019)
	Indirect N2O from NH3	0.01	0.007	0.013	kg N2O-N/NH3-N	+/-30%, (Miljødirektoratet 2019)

Table S9: Characterization factors used for Near-term Climate Forcers (NTCF) for GWP20 and GWP100 taken from (Levasseur et al. 2016). VOC: volatile organic carbon; OC: organic carbon. Characterization factor for NOx are given on a nitrogen basis in (Levasseur et al. 2016), and were converted to NOx basis assuming that NOx corresponds to a 50%/50% share of NO and NO_2, (i.e. average molecule of $NO_{1.5}$.). No range were available for SO_x.

Metric	NTCF	Baseline	Low bound	High bound	Unit
GWP20, Global	NOx	-40	-53	-27	kg CO2eq/kg NOx
	CO	7.8	5.8	9.8	kg CO2eq/kg CO
	VOC	18.7	11.2	26.2	kg CO2eq/kg VOC
	OC	-160	-320	-60	kg CO2eq/kg OC
	Black carbon	3200	270	6200	kg CO2eq/kg BC
	SOx	-141	n.a.	n.a.	kg CO2eq/kg SOx
GWP100, Global	NOx	-11	-14	-7.4	kg CO2eq/kg NOx
	CO	2.1	1.6	2.6	kg CO2eq/kg CO
	VOC	5.5	3.2	7.8	kg CO2eq/kg VOC
	OC	-43	-86	-17	kg CO2eq/kg OC
	Black carbon	846	94	1600	kg CO2eq/kg BC
	SOx	-38	n.a.	n.a.	kg CO2eq/kg SOx

Table S10: Parameters and ranges of values considered for the uncertainty analysis

	Parameter	Baseline	Low bound	High bound	Unit	Comments
Barley yield	In reference and biochar scenarios	3756	3266	4246	kg barley/ha	See section 2.2 in main text
	In biochar-fertilizer scenarios	4545	3643	5296	kg barley/ha	See section 2.7 in main text
Biochar's effect in soils	N2O from fertilizer	-38	-50	-22	%	See section 2.7 in main text; percentage change compared to values in Table S1
	NOx from fertilizer	-10	-20	0	%	See section 2.7 in main text; percentage change compared to values in Table S1
	NH3 from fertilizer	5	0	10	%	See section 2.7 in main text; percentage change compared to values in Table S1
	N leaching	-8	-16	0	%	See section 2.7 in main text; percentage change compared to values in Table S1
Distribution of PM to BC and OC from life cycle value chain	Organic carbon	0.09	0.007	0.18	Mass fraction of PM10	Baseline: middle of range Variability: taken from (Bond et al. 2004)
	Black carbon	0.30	0.023	0.57	Mass fraction of PM10	Baseline: middle of range Variability: taken from (Bond et al. 2004)
Pyrolysis and biochar	Biochar yield dry basis	0.285	0.242	0.327	kg biochar/kg feedstock	Baseline from our modeling of pyrolysis (mass balance) Variability: +/-15% (assumption)
	Pyrolysis duty	0.08	0.06	0.15	MJ/MJ in feedstock	Baseline taken from Aspen simulation Variability: (Crombie and Mašek 2014)
	Biochar C content	79	71.1	86.9	%	Baseline from our modeling of pyrolysis (mass balance) Variability: +/-10% (assumption)
	Biochar C stability in soils	74	63	80	%	Baseline taken from (Budai et al. 2016) for Norwegian conditions

						Varibility: low bound average from (Budai et al. 2016); high bound IPCC value for biochar produced above 450°C
	Bio-oil C content	61	54.9	67.1	%	Baseline from our modeling of pyrolysis (mass balance) Variability: +/-10% (assumption)
Transport	Forest to biochar plant	190	152	228	km	Baseline, see main text Variability: +/-20% (assumption)
	Biochar plant to field	226	181	271	km	Baseline, see main text Variability: +/-20% (assumption)

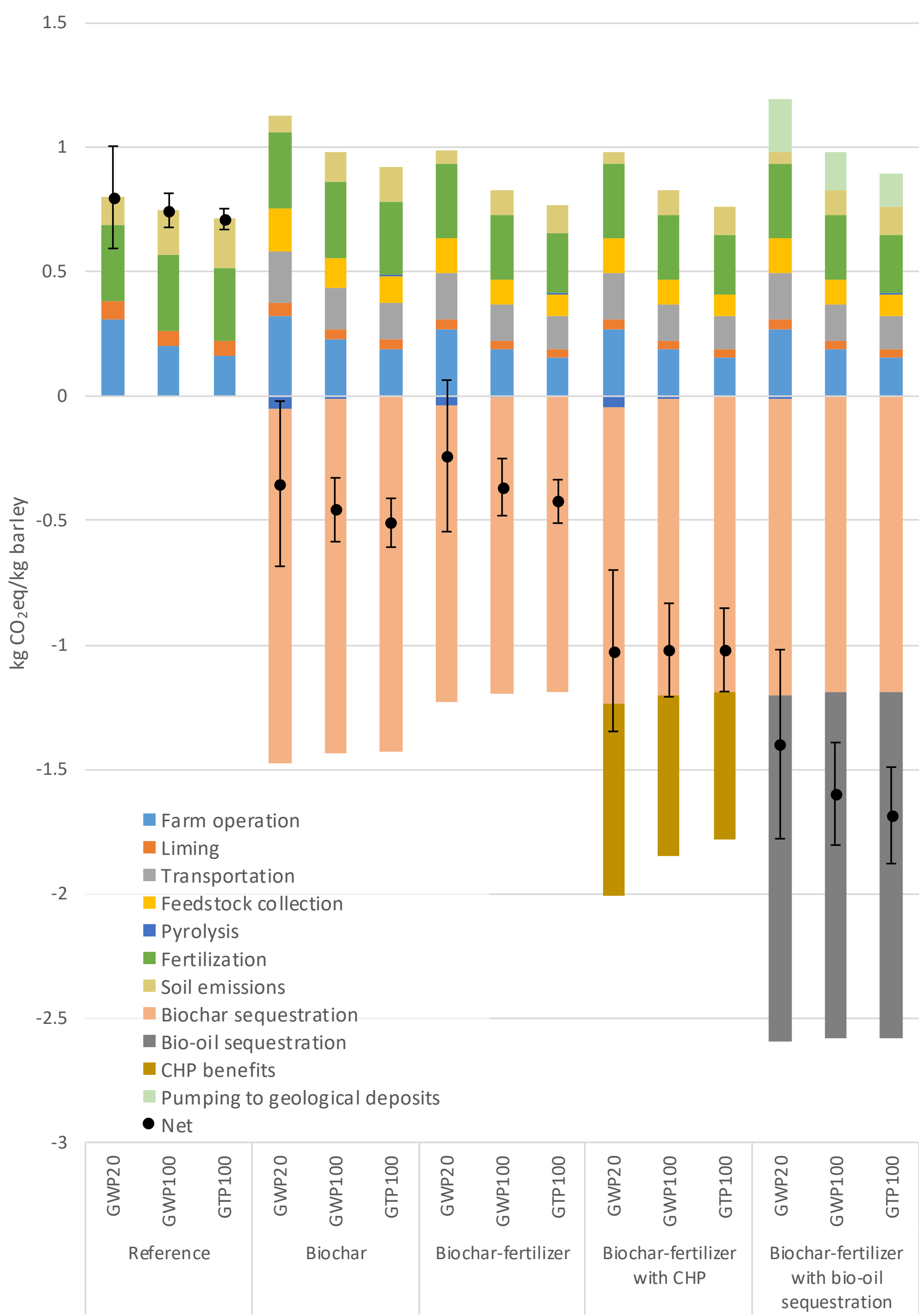

Figure S6: Climate change effects of the five scenarios using different metrics for characterization of impacts: global warming potential at 20 years' time horizon (GWP20), global warming potential at 100 years' time horizon (GWP100) and global temperature potential at 100 years' time horizon (GTP100). Results are presented on kg barley basis. Black dots represent the net impact and the whiskers show uncertainty range from our Monte-Carlo analysis (± one standard deviation).

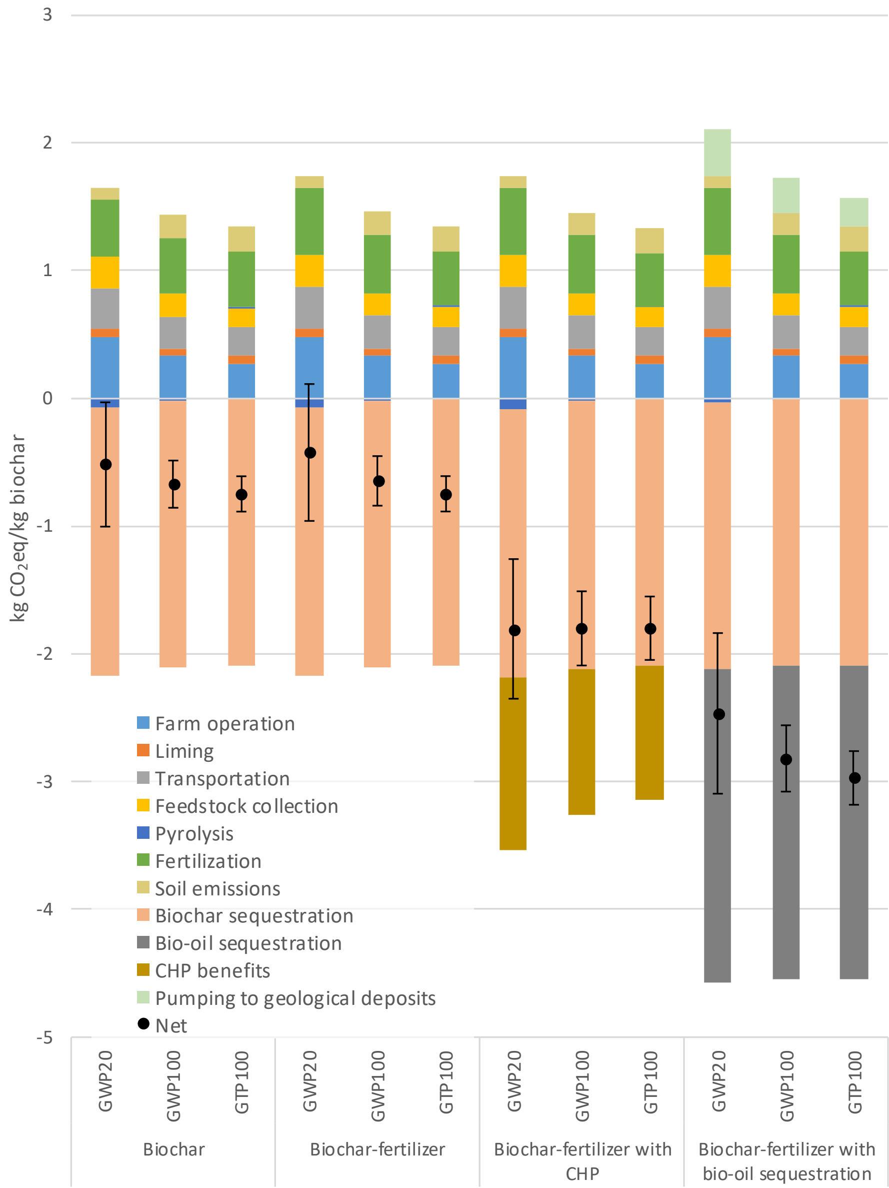

Figure S7: Climate change effects of the five scenarios using different metrics for characterization of impacts: global warming potential at 20 years' time horizon (GWP20), global warming potential at 100 years' time horizon (GWP100) and global temperature potential at 100 years' time horizon (GTP100). Results are presented on kg biochar basis. Black dots represent the net impact and the whiskers show uncertainty range from our Monte-Carlo analysis (± one standard deviation).

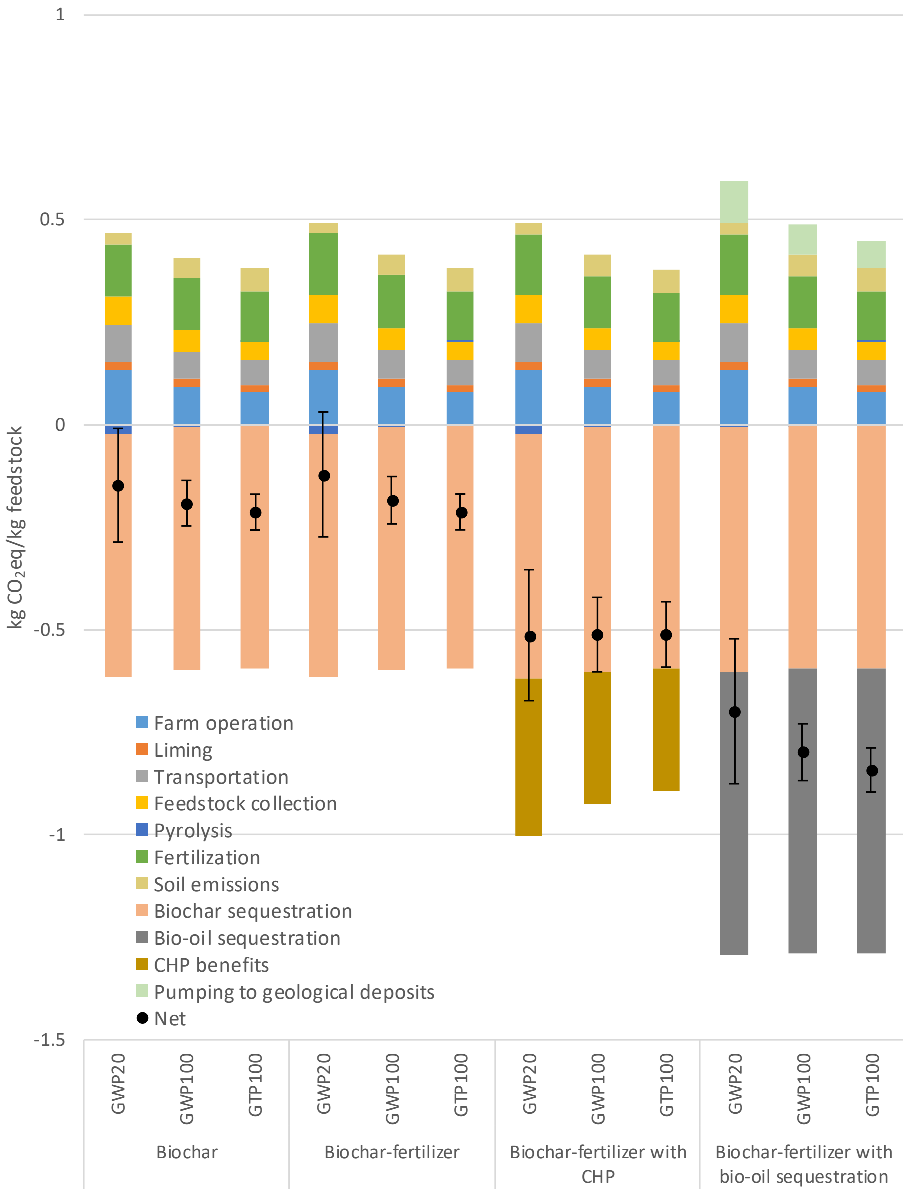

Figure S8: Climate change effects of the five scenarios using different metrics for characterization of impacts: global warming potential at 20 years' time horizon (GWP20), global warming potential at 100 years' time horizon (GWP100) and global temperature potential at 100 years' time horizon (GTP100). Results are presented on kg feedstock basis. Black dots represent the net impact and the whiskers show uncertainty range from our Monte-Carlo analysis (± one standard deviation).

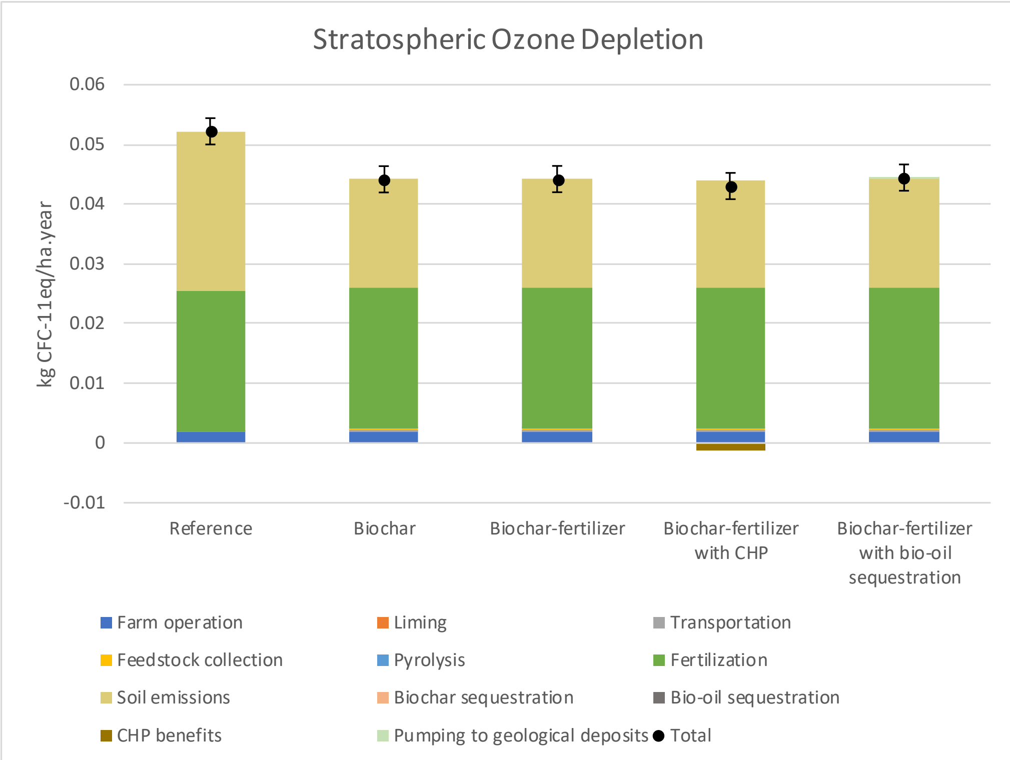

Figure S9: Stratospheric ozone depletion effects of the biochar scenarios against a reference system. Transportation accounts for both feedstock and biochar. Black dots represent the net climate impact and the whiskers show uncertainty ranges from the Monte-Carlo analysis (± one standard deviation).

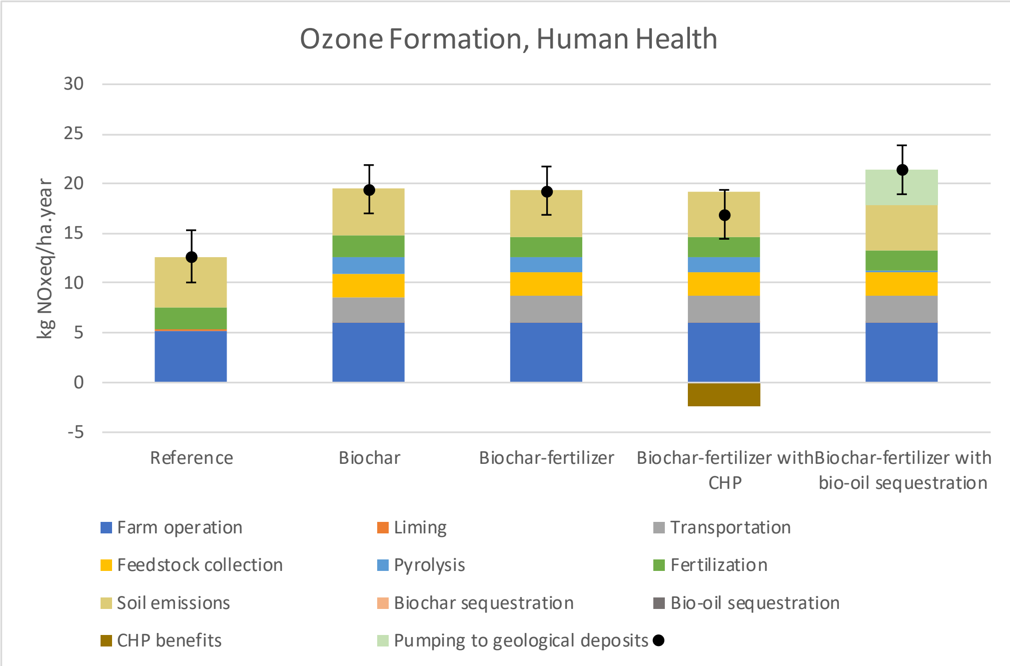

Figure S10: Ozone formation (human health) effects of the biochar scenarios against a reference system. Transportation accounts for both feedstock and biochar. Black dots represent the net climate impact and the whiskers show uncertainty ranges from the Monte-Carlo analysis (± one standard deviation).

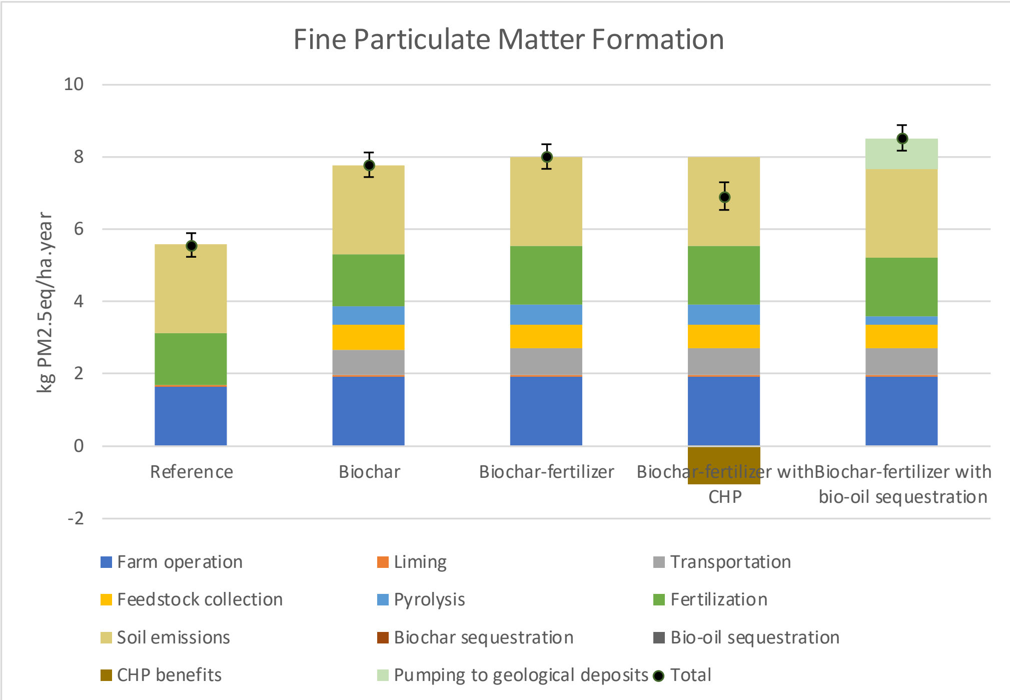

Figure S11: Fine particulate matter effects of the biochar scenarios against a reference system. Transportation accounts for both feedstock and. Black dots represent the net climate impact and the whiskers show uncertainty ranges from the Monte-Carlo analysis (± one standard deviation).

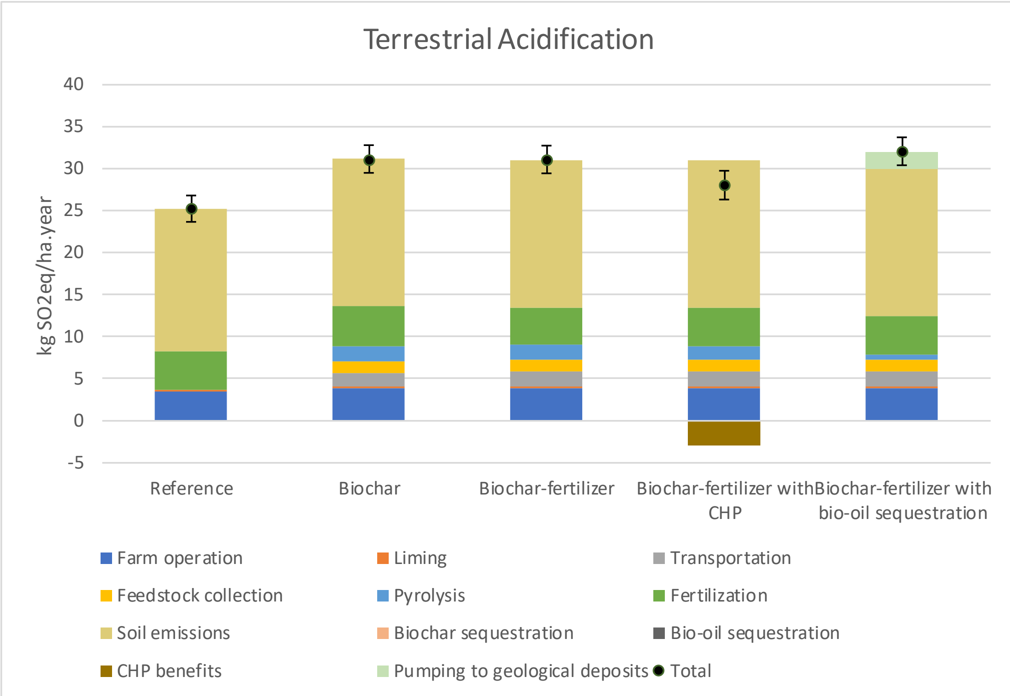

Figure S12: Terrestrial acidification effects of the biochar scenarios against a reference system. Transportation accounts for both feedstock and biochar. Black dots represent the net climate impact and the whiskers show uncertainty ranges from the Monte-Carlo analysis (± one standard deviation).

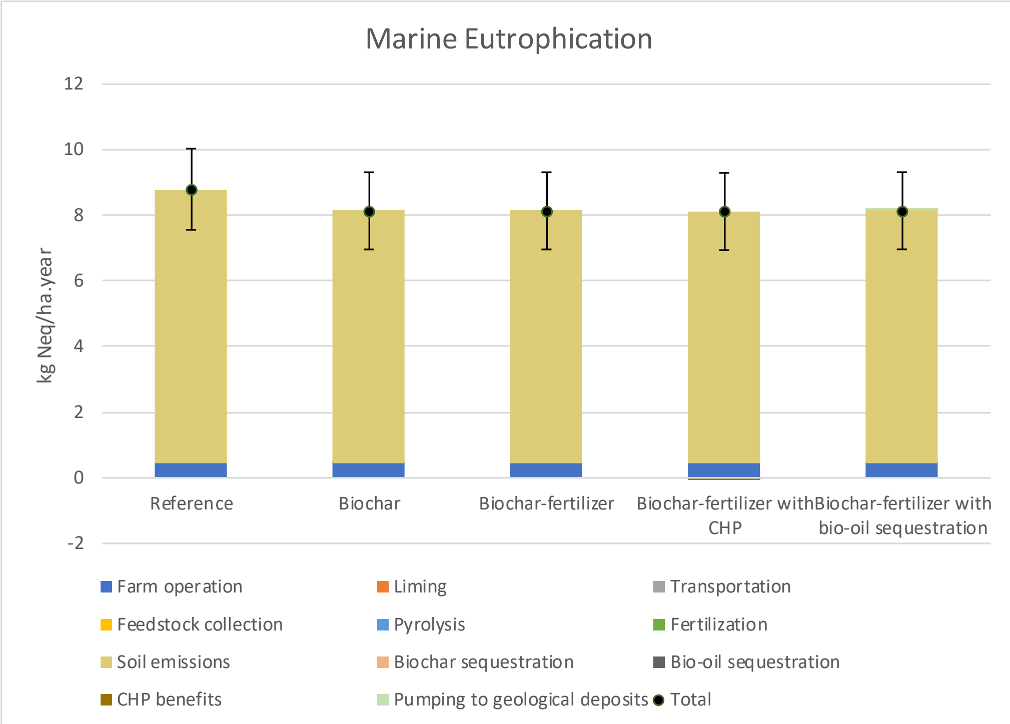

Figure S13: Marine eutrophication effects of the biochar scenarios against a reference system. Transportation accounts for both feedstock and biochar. Black dots represent the net climate impact and the whiskers show uncertainty ranges from the Monte-Carlo analysis (± one standard deviation).

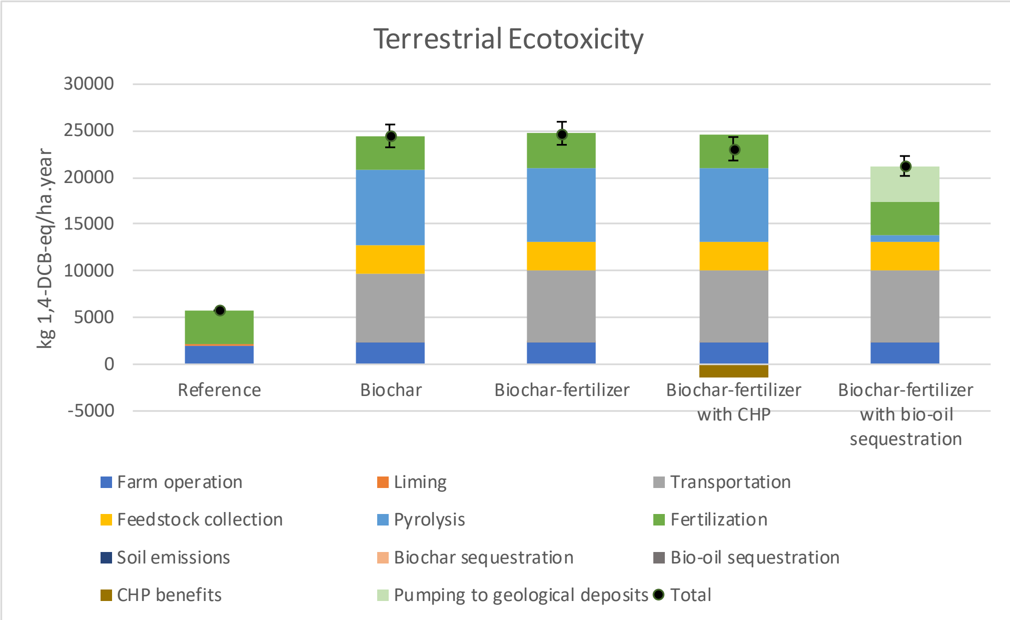

Figure S14: Terrestrial ecotoxicity effects of the biochar scenarios against a reference system. Transportation accounts for both feedstock and biochar. Black dots represent the net climate impact and the whiskers show uncertainty ranges from the Monte-Carlo analysis (± one standard deviation).

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6.3 Supplementary information to Chapter 4

Supplementary Information: Biochar and its potential to deliver negative emissions and better soil quality in Europe

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Table 1: Combinations of different supply potentials for crop and forest residues. For each case, the table shows the amount of currently used residues, the biochar potential, the area treated each year at 5 or 30 t/ha and the number of years needed to treat all cropland in Europe (168 Mha) at the corresponding residue availability. The potentials considered in our study are given by the following combinations: sustainable potential for crop residues and base potential for the "low residue supply potential", with subtracted volumes of used residues; Technical potential for crop residues and high potential for forest residues for the "high residue supply potential", without subtracting volumes of used residues. The scenarios of availability of crop residues is from ref. [1], and of forest residues from ref. [2].

Crop residues potentials (Mtonnes)		Forest residues potentials (Mtonnes)			Used residues (Mtonnes)		Biochar potential (Mtonnes)	Area treated per year (Mha)		Time to treat all cropland (years)	
Sustainable	Technical	BASE	HIGH	TECH	Crop	Forest		5 tonnes biochar/ha	30 tonnes biochar/ha	5 tonnes biochar/ha	30 tonnes biochar/ha
149	-	39	-	-	-29	-32	30.6	6.1	1.0	28	166
149	-	-	75	-	-29	-32	40.0	8.0	1.3	21	127
149	-	-	-	105	-29	-32	47.8	9.6	1.6	18	107
-	212	39	-	-	-29	-32	45.7	9.1	1.5	19	111
-	212	-	75	-	-29	-32	55.1	11.0	1.8	15	92
-	212	-	-	105	-29	-32	62.9	12.6	2.1	13	81
149	-	39	-	-	-	-	45.9	9.2	1.5	18	111
149	-	-	75	-	-	-	55.3	11.1	1.8	15	92
149	-	-	-	105	-	-	63.1	12.6	2.1	13	81
-	212	39	-	-	-	-	61.0	12.2	2.0	14	83
-	212	-	75	-	-	-	70.4	14.1	2.3	12	72
-	212	-	-	105	-	-	78.2	15.6	2.6	11	65

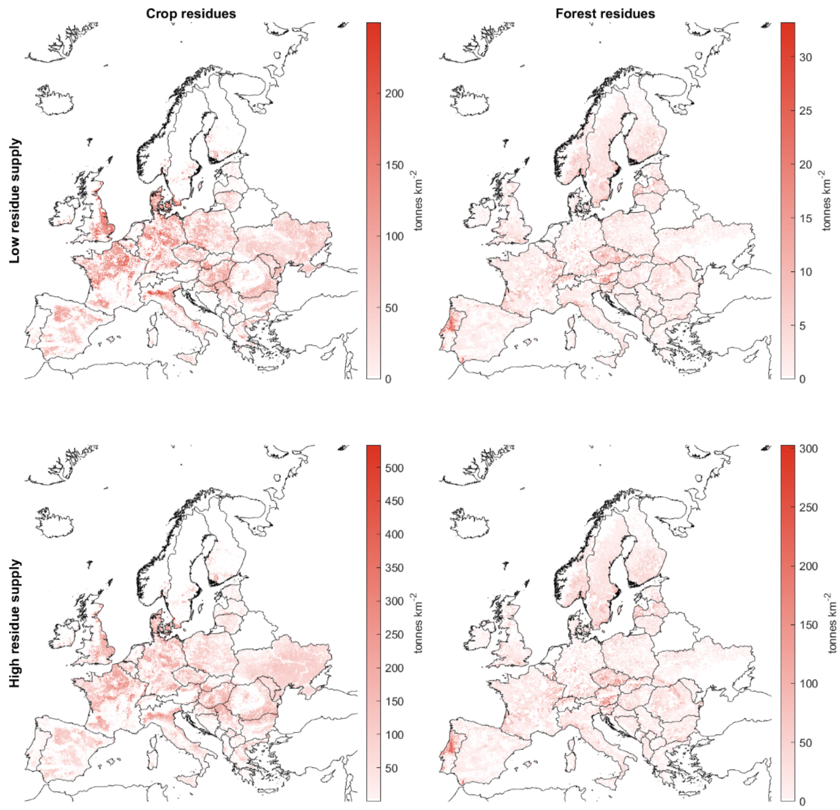

Figure 1: Spatial distribution of crop and forest residues availability in Europe. The map has a resolution of 1 km².

Table 2: Scale of deployment of pyrolysis technology under different residues supply potentials and size of pyrolysis plants

Crop residues potentials (Mtonnes)		Forest residues potentials (Mtonnes)			Used residues (Mtonnes)		Plant capacity (ktonnes feedstock/year)
Sustainable	Technical	BASE	TECH	HIGH	Crop	Forest	560
							Number of biochar plants
149		38			-29	-32	226
149			71		-29	-32	286
149				105	-29	-32	347
	212	38			-29	-32	340
	212		71		-29	-32	399
	212			105	-29	-32	460
149		38			0	0	336
149			71		0	0	395
149				105	0	0	456
	212	38			0	0	449
	212		71		0	0	509
	212			105	0	0	570

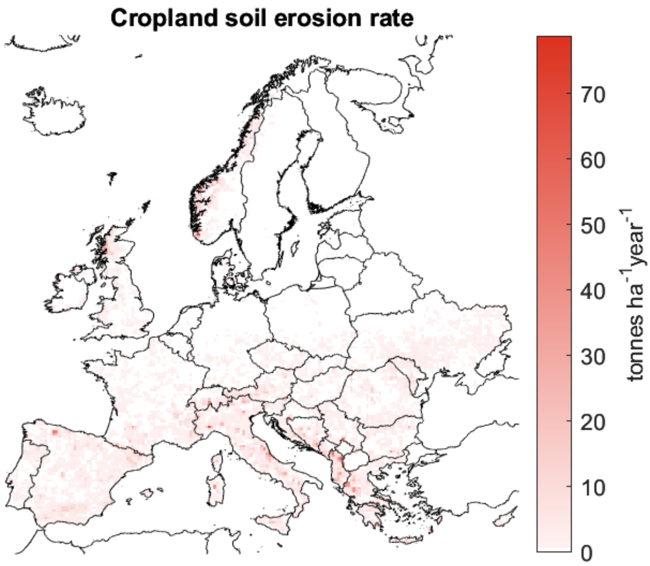

Figure 2: Map of erosion rates in European cropland.

Table 3: Summary statistics of tranport distances for different biomass materials estimated with our logistic models under two residue supply potentials.

		min	10 %	25 %	median	75 %	90 %	max	mean
Low residues supply	Residues transport	27	37	44	53	62	76	145	56
	Bio-oil transport	28	519	810	1253	1817	2346	2802	1343
	Biochar transport (5t/ha)	8	19	27	41	64	119	655	59
	Biochar transport (30 t/ha)	8	11	24	40	94	197	1670	90
High residues supply	Residues transport	27	27	33	42	51	62	226	44
	Bio-oil transport	28	542	862	1313	1870	2343	2872	1377
	Biochar transport (5t/ha)	8	19	27	42	100	447	1787	140
	Biochar transport (30 t/ha)	8	11	21	34	62	141	1395	66

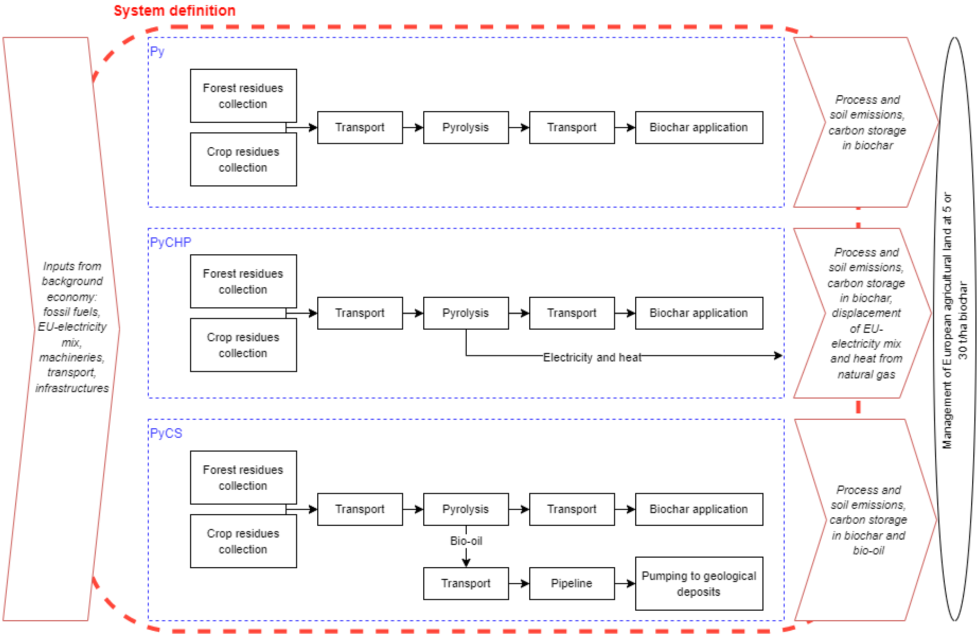

Figure 3: System boundaries and main stages of the LCA.

Table 4: Characterization factors used for Near-term Climate Forcers (NTCFs) for GTP20 and GWP100. NMVOC: non-methane volatile organic carbon; OC: organic carbon. Taken from Aamaas et al.[3]

Metric	Climate forcer	Baseline	Low bound	High bound	Unit
GTP20, Europe	NOx	-24	-38	-9.2	kg CO2eq/kg NOx
	NH3	-13	-16	-7	kg CO2eq/kg NH3
	CO	4.6	2.5	6.7	kg CO2eq/kg CO
	NMVOC	18.5	-5.6	46	kg CO2eq/kg VOC
	OC	-165	-377	-42	kg CO2eq/kg OC
	Black carbon	575	436	810	kg CO2eq/kg BC
	SOx	-83	-206	-22	kg CO2eq/kg SOx
	CH4	48	36	66	kg CO2eq/kg CH4
GWP100, Europe	NOx	-10	-15.8	-3.6	kg CO2eq/kg NOx
	NH3	-13	-15	-6.7	kg CO2eq/kg NH3
	CO	2.3	1.2	3.5	kg CO2eq/kg CO
	NMVOC	10	-7	28	kg CO2eq/kg VOC
	OC	-165	-354	-39	kg CO2eq/kg OC
	Black carbon	540	405	760	kg CO2eq/kg BC
	SOx	-77	-193	-22	kg CO2eq/kg SOx
	CH4	23	17	33	kg CO2eq/kg CH4

Table 5: Parameters and ranges of values considered for the uncertainty analysis.

	Parameter	Baseline	Low bound	High bound	Unit	Comments
LCA emission factors		1	0.8	1.2	-	Scaling factor Variability: +/-20% (assumption)
Distribution of PM to BC and OC from life cycle value chain	Organic carbon	0.09	0.007	0.18	Mass fraction of PM10	Baseline: middle of range Variability: taken from Bond et al.[4]
	Black carbon	0.30	0.023	0.57	Mass fraction of PM10	Baseline: middle of range Variability: taken from Bond et al.[4]
Pyrolysis and biochar	Straw biochar yield dry basis	0.24	0.192	0.288	kg biochar/kg feedstock	Baseline: see main text Variability: +/-20% (assumption)
	Wood biochar yield dry basis	0.26	0.208	0.312	kg biochar/kg feedstock	
	Biochar C content	82	69.7	94.3	%	Baseline from our modeling of pyrolysis (mass balance) Variability: +/-15% (assumption)
	Biochar C stability in soils	80	71.2	88.8	%	IPCC value for biochar produced above 450°C, 95% confidence interval
	Bio-oil C content from wood	64	51.2	76.8	%	Equation from Woolf et al. (2014)[5] Variability: +/-10% (assumption)
	Bio-oil C content from straw	61	48.8	73.2	%	Equation from Woolf et al. (2014)[5] Variability: +/-10% (assumption)
Transport		1	0.8	1.2	-	Scaling factor for the transport distances Variability: +/-20% (assumption)

Table 6: Uncertainty ranges for biochar's effects on soils

	Parameter	Baseline	Low bound	High bound	Unit	Comments
Biochar's effect on soils	Crop yield	Grid cell	-5.4 (-5.8)	5.4 (5.8)	%	Liu et al.[6]
	N_2O emissions	Grid cell	-9.5 (-7)	9.5 (7)	%	
	NH_3 emissions	Grid cell	-28.5 (-20)	28.5 (20)	%	
	Nleach	Grid cell	-11 (-7)	11 (7)	%	
	NO_x	-10 (-34)	-20 (-67)	0 (0)	%	See main text
	WHC	Grid cell	-15 (-7)	15 (7)	%	Baseline model from

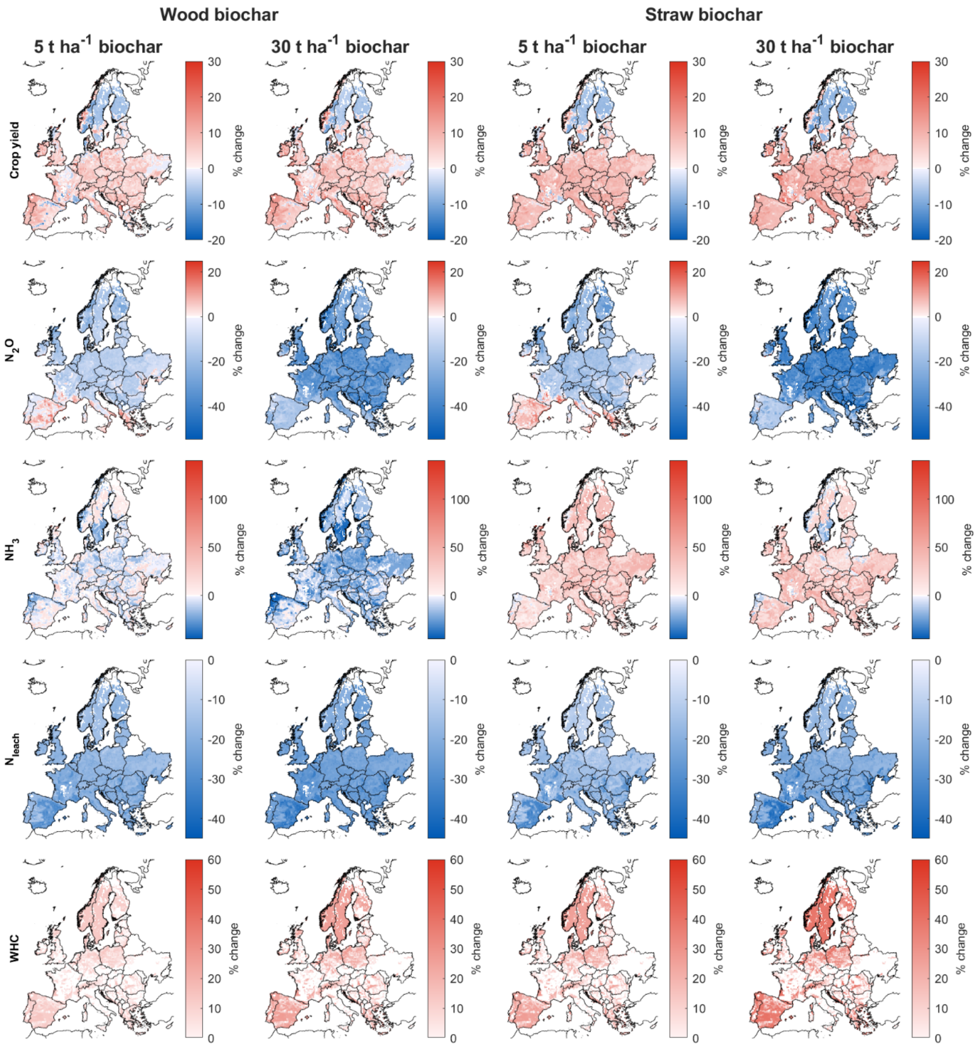

Figure 4: Predicted effects of wood and straw biochar on soils at application rates of 5 or 30 t ha-1. Data for crop yield, N_2O, NH_3 and nitrogen leaching are from Liu et al.[6], data for water holding capacity (WHC) are based on the empirical model from Kroeger et al.[7]

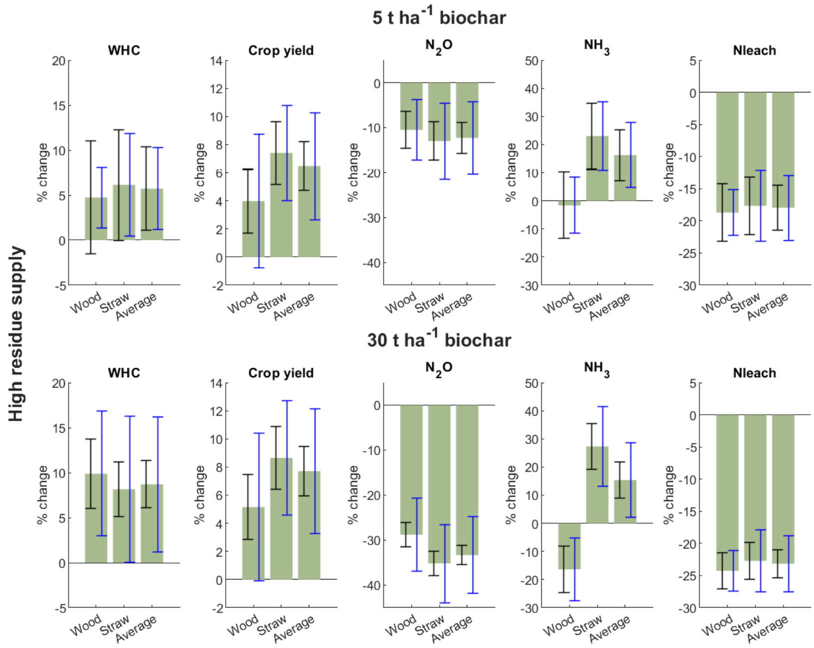

Figure 5: Effects of biochar on soil water holding capacity (WHC), crop yield, N_2O and NH_3 emissions, and nitrogen leaching averaged across European cropland over 30 years. Results are shown for both wood and straw biochar, and the average effect of the two. Black whiskers represent the uncertainty of the mean effect (± standard deviation) from a Monte-Carlo analysis based on the uncertainty in the response of each individual grid, while the blue whiskers represent the mean ± standard deviation of the spatial variability of the effect.

Supplementary text 1 – Biochar effects on soils

Based on a meta-analysis, biochar may have only transient effects on soil N_2O emissions, disappearing after one year[8]. Biochar's effect on soil NO_x emissions is less studied N_2O, and its long-term effect have not been investigated. As soil NO_x emissions are usually correlated to N_2O, the effect may be also only transient[9]. Biochar can increase NH_3 volatilization due to its alkalinity, moving chemical equilibrium towards NH_3, while its surface chemistry can help retain NH_4^+ and help reduce its volatilization as NH_3[10]. For these reasons, the increase in NH_3 can be expected to be transient, as oxidation of its surface will help to better retain NH_4^+ and its alkalinity will decline[10]. However, biochar is also advocated to reduce soil bulk density and compaction, thus increasing soil aeration. In this case, the increase in NH_3 volatilization due to biochar could be sustained, if diffusion is the limiting factor to NH_3 volatilization. Biochar can retain nitrate in soils, but it is unclear as how it will develop over years once the mixture of soil and biochar reaches its new maximal capacity. It will likely depend on nitrogen cycling in soil, uptake by plants and input of nitrogen fertilizer.

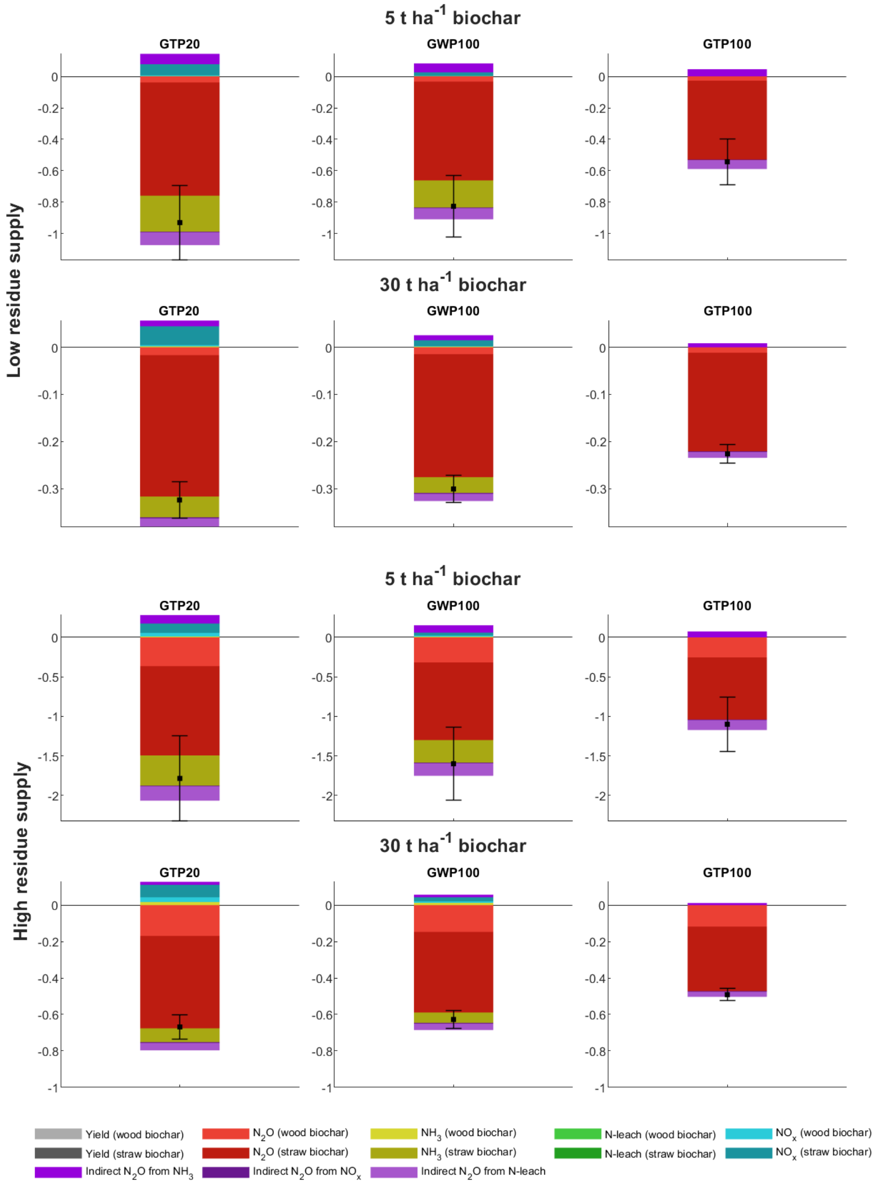

Figure 6: Climate change impacts of soil emissions using three climate change indicator (GTP20, GWP100, and GTP100) representative of a short, medium and long term response of the climate system (see Methods). Black whiskers are the mean ± standard deviation of the change in climate impacts from soil emissions at the European continental scale, taking into account the uncertainties in biochar's effects to soil emissions and of the characterization factors. In the shorter-term (GTP20), mitigation is stronger as the climate forcing of several near-term climate forcers (and N_2O) is higher. In the long term (GTP100), the mitigation potential is smaller because the effects of near-term climate forcers (mainly NH_3 and NO_x) become negligible, and only contributions from the long-lived gas N_2O remain.

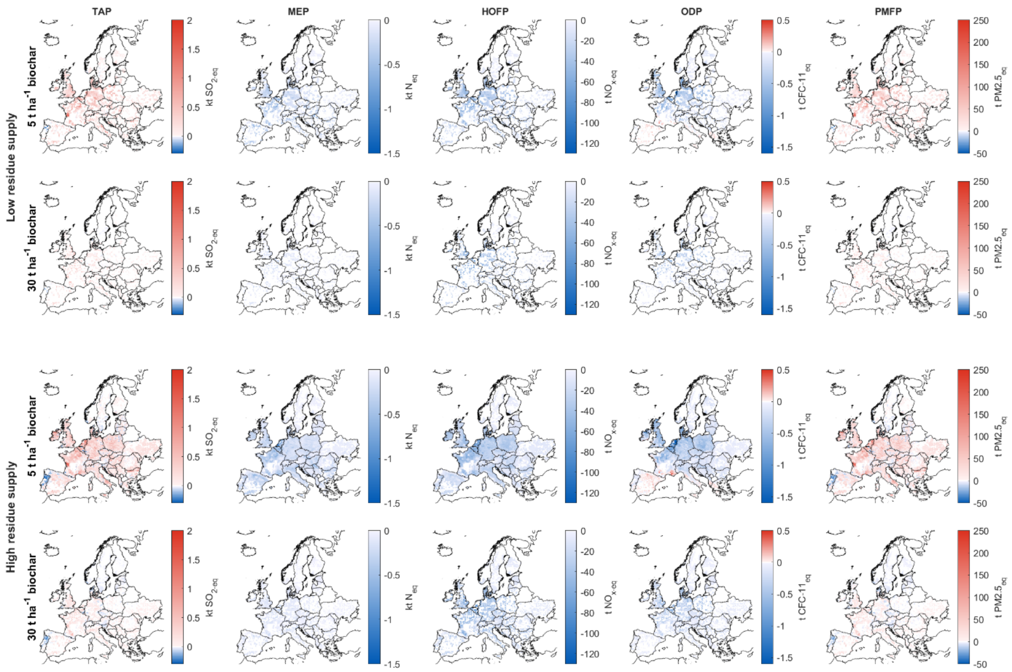

Figure 7: Grid contribution to changes in environmental impacts from soil emissions. TAP: terrestrial acidification potential, MEP: marine eutrophication potential, HOFP: tropospheric ozone formation potential, ODP: stratospheric ozone depletion potential and PMFP: particulate matter formation potential. Results are shown for biochar produced from two residue supply cases (low and high) and for two different biochar application rates (5 and 30 t ha^{-1}) to agricultural soils in Europe (treating first those most threatened by soil erosion).

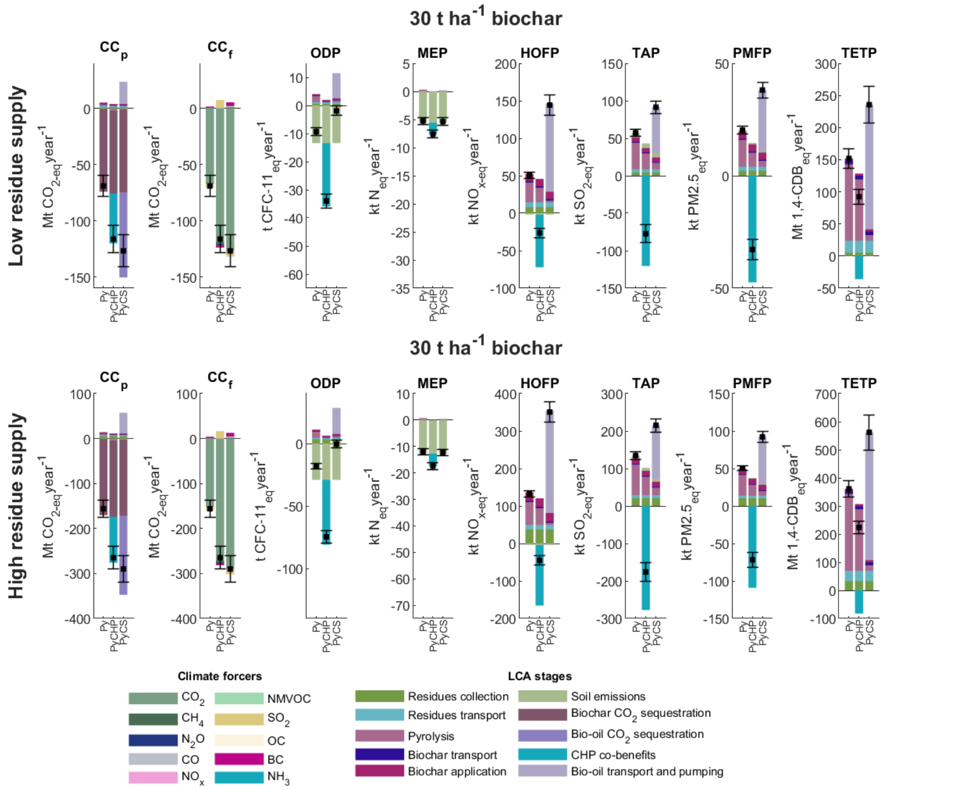

Figure 8: Annual average total impacts from a life-cycle perspective integrating direct and indirect emissions from the biochar systems with biochar-induced soil emissions. Results are shown for two residue supply scenarios (low and high) and a biochar application rate to agricultural soils of 30 t ha^{-1}. The three biochar technologies considered are: Py, biochar only, with no external benefits from pyrolysis co-products; PyCHP, biochar with CHP, with the co-products of the pyrolysis used in a CHP system to generate electricity (replacing European electricity mix) and heat (replacing heat produced from natural gas); PyCS, biochar and bio-oil sequestration, with the bio-oil produced during pyrolysis is recovered, transported by truck and pumped to geological deposits for storage. Impacts are climate change per process (CCp) and climate forcers (CCf), terrestrial acidification (TAP), marine eutrophication (MEP), tropospheric ozone formation (HOFP), stratospheric ozone depletion (ODP), fine particulate formation (PMFP) and terrestrial ecotoxicity (TETP). Black points represent the mean net effect and whiskers show one standard deviation. Note: different axis scale between low and high residues supply.

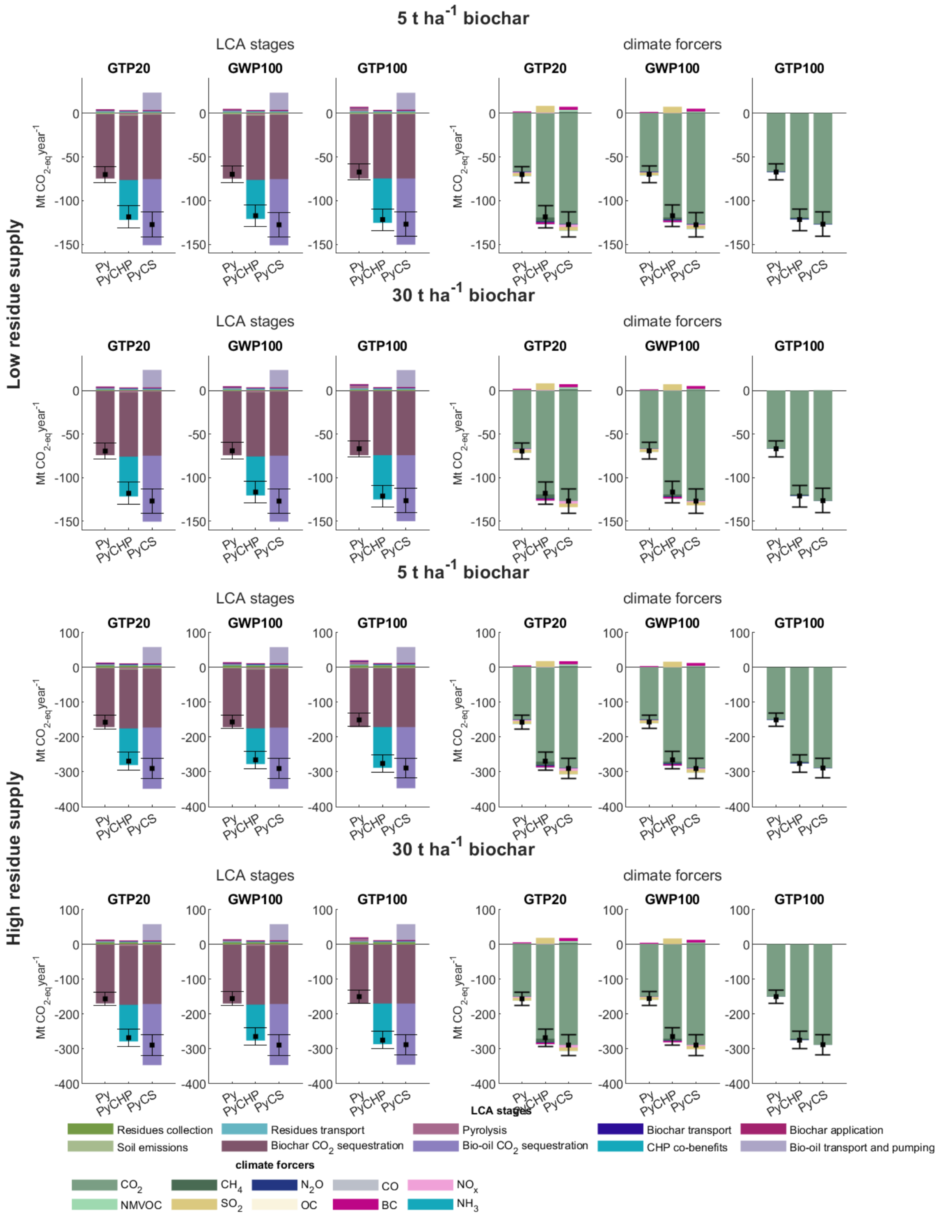

Figure 9: Annual average total climate impacts from a life-cycle perspective using three climate metrics. GTP20: global temperature potential 20 years, GWP100: global warming potential at 100 years time horizon, GTP100: global temperature potential in 100 years. These indicators represent a short, medium and long term response of the climate system (see Methods). Results integrate direct and indirect emissions from the biochar systems with biochar-induced soil emissions and are shown for two residue supply potentials (low and high) and a biochar application rate to agricultural soils of 5 t ha^{-1}. The three biochar technologies considered are: Py, biochar only, with no external benefits from pyrolysis co-products; PyCHP, biochar with CHP, with the co-products of the pyrolysis used in a CHP system to generate electricity (replacing European electricity mix) and heat (replacing heat produced from natural gas); PyCS, biochar and bio-oil sequestration, with the bio-oil produced during pyrolysis is recovered, transported by truck and pumped to geological deposits for storage.

Supplementary text 2 – Uncertainty and limitations

Biochar's effect on soil NOx emissions were not spatially explicit due to limited available data. Soil emissions of NOx can happen during denitrification and nitrification processes in the soil and are correlated to N2O emissions[9]. There is a possibility that biochar increases soil NOx emissions in some European regions, maybe following biochar's effect on N2O emissions pattern.

There are other possible benefits of biochar that have not been explicitly considered. Biochar can help reduce soil losses by increasing wet soil aggregation[11] and retaining water[12], particularly in coarse soils. In fine soils, saturated hydraulic conductivity tend to increase[12], improving water percolation in the soil and retarding run-offs[13]. However, only limited data are available on biochar's effect on soil erosion and contrasting findings across locations and soil types are reported[14,15].

While increase in WHC is usually observed in coarse soils (and was modelled only for soils with sand content over 45%, see Methods), biochar can also improve other hydrological properties of soils in other texture classes such as increased plant available water[12], and increased saturated hydraulic conductivity in fine textured soil, which would improve water penetration in soil and reduce run-offs in such soils[12].

One effect of biochar in soils that was not modelled in this study is its capacity in stabilizing native soil organic carbon, a process called negative priming. Reducing the degradation rate of native soil organic carbon could allow to further increase carbon stock in soils after biochar incorporation. While the effect is mostly a reduction in priming over long-term, a positive priming can also be observed in the short-term and the scale of the effect is dependent on soil conditions and biochar properties[16]. The effect can be important, for example a study found that under no-till conditions accumulation of non-biochar carbon in soil was about twice the initial amount of carbon incorporated after a six year period[17]. A proper understanding of this effect and modeling approach to estimate the new soil carbon equilibrium after biochar incorporation is required before being able to include this effect in the quantification of climate mitigation potential of biochar.

Climate change effects can also be affected by a potential decline in soil organic carbon stocks of forest areas because of the removal of a share of residues. Agricultural residues used for biochar are returned to agricultural soils, but forest residues are diverted from forests to cropland. Forest residues naturally oxidize to CO_2 over time, but part of it would return to soils. In a study based on Nordic conditions[18], soil carbon declines for a couple of decades, but then it stabilizes and in the long-term the cumulative loss of soil carbon in the forest is about 10% of the corresponding biochar-induced increase in agricultural land. Further, the carbon stored in the form of biochar is more stable than the litter carbon from forest residues, so representing a safer option for a long-term carbon storage. Another challenge for forest soils is the potential removal of nutrients with residues, especially nitrogen, whose limited availability can be a constrain to forest growth in some locations[19]. To prevent an additional use of fertilizers, an option to limit nutrient removal with residues is to leave them on the forest floor for one year before collection, so that branches have time to lose foliage and needles (which store about 50% of the N content of the residues).

Effects of biochar on availability of heavy metals and pesticides (which would affect terrestrial ecotoxicity) are excluded but they are expected to play a relatively minor role in the impact assessment carried out through a life-cycle assessment perspective. It is unlikely that a reduction in the availability of heavy metals in soils can offset the effect on terrestrial ecotoxicity, which is primarily linked to the emissions of heavy metals in the supply chain of biochar. In a previous study, pesticides had a negligeable contribution to terrestrial ecotoxicity impacts[20].

Our analysis relies on large-scale biochar production, with a plant capacity of about 560 ktonnes feedstock, which is large compared to current planned or in operation bio-energy plants in Europe that typically treat up to 250-350 ktonnes residues (https://demoplants.best-research.eu/). However, the modeling approach specified here represents a maximum installed capacity based on techno-economic optimization. A more decentralized biochar production with smaller plant capacity would reduce transport needs for both residues and biochar, thus not negatively impacting our results (especially since transport is already a small contributor to most of the impacts). At the same time, larger pyrolysis units could implement better emissions control and reduce the effects on air pollution highlighted in our study. For example, pyrolysis was the main, large, contributor to terrestrial ecotoxicity impacts. Impacts were adapted from the study of a commercially available small-scale pyrolyser[21], and these emissions could be considerably reduced by installing a scrubber or filter as heavy metals are associated with the particulate matter. Future decarbonization of the energy system will decrease the environmental benefits of the CHP case, because the electricity from the grid will likely rely more on renewable energy and less on fossil energy. At the same time, the large contribution to warming emissions due to bio-oil transport for sequestration could be reduced by using other transport means such as rail freight. Storing of bio-oil into geological deposits needs to be further studied as it could face technical challenges and limitations. It generally has high viscosity and is corrosive, which could make its handling, storage and transport difficult. It also contains toxic compounds and would require careful handling[22]. However a recent study argue that bio-oil can have pumping and transport properties similar to crude oil[23]. If bio-oil sequestration turns out unfeasible, it could also be upgraded to biofuels displacing fossil fuels with additional climate mitigation benefits, or mixed with asphalt for road construction, which would represent another storage of carbon[24].

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