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Evaluating climate change mitigation potential of hydrochars: compounding insights from three different indicators

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Abstract

We employed life cycle assessment to evaluate the use of hydrochars, prospective soil conditioners produced from biowaste using hydrothermal carbonization, as an approach to improving agriculture while using carbon present in the biowaste. We considered six different crops (barley, wheat, sugar beet, fava bean, onion, and lucerne) and two different countries (Spain and Germany), and used three different indicators of climate change: global warming potential (GWP), global temperature change potential (GTP), and climate tipping potential (CTP). We found that although climate change benefits (GWP) from just sequestration and temporary storage of carbon are sufficient to outweigh impacts stemming from hydrochar production and transportation to the field, even greater benefits stem from replacing climate-inefficient biowaste management treatment options, like composting in Spain. By contrast, hydrochar addition to soil is not a good approach to improving agriculture in countries where incineration with energy recovery is the dominant treatment option for biowaste, like in Germany. Relatively small, but statistically significant differences in impact scores (ISs) were found between crops. Although these conclusions remained the same in our study, potential benefits from replacing composting were smaller in the GTP approach, which due to its long-term perspective gives less weight to short-lived greenhouse gases (GHGs) like methane. Using CTP as indicator, we also found that there is a risk of contributing to crossing of a short-term climatic target, the tipping point corresponding to an atmospheric GHG concentration of 450 ppm CO_2 equivalents, unless hydrochar stability in the soil is optimized. Our results highlight the need for considering complementary perspectives that different climate change indicators offer, and overall provide a foundation for assessing climate change mitigation potential of hydrochars used in agriculture.

Keywords: biowaste, climate tipping, global temperature change potential, hydrothermal carbonization, life cycle assessment, waste management

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Introduction

Hydrochar is a carbonaceous material produced from biomass residues using hydrothermal carbonization (HTC; Berge *et al.*, 2011; Titirici *et al.*, 2014). It is mainly used as solid fuel for domestic heating, but its use in agriculture as soil conditioner with some carbon storage value has recently attracted attention (Reza *et al.*, 2014; Burguete *et al.*, 2016). Hydrochar has similar properties to pyrolytic biochar, although the presence of water and lower process temperature (180–250 °C) (when compared to dry pyrolysis) make hydrochar less stable in the soil compared to pyrolytic biochar. Recent studies investigated various aspects of hydrochar use for crop production, including its influence on seed germination, plant morphology, crop productivity, or nutrient release from the hydrochar to the soil (e.g., Malghani *et al.*, 2014; Reibe *et al.*, 2014; Schimmelpfennig *et al.*, 2015). Because of the yet insufficient amount of data on these aspects, more research was needed to determine effects of hydrochar on crop production and soil processes (Reza *et al.*, 2014).

Assessment of environmental performance of hydrochars, including assessment of their potential contribution to climate change mitigation, can be quantified using life cycle assessment (LCA). In LCA, resource consumption and emissions of pollutants stemming from the extraction of the raw materials (e.g., for HTC plant), their manufacture and use or operations (e.g., for

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running the plant) up to their end of life (e.g., disposal of post-treatment ashes and recycling operations) are inventoried. These life cycle inventories are then translated into impact indicator scores using substance-specific characterization factors for various life cycle impact categories, like climate change (Hauschild, 2005; Hellweg & Mila i Canals, 2014). Studies investigating environmental performance of hydrochars using LCA have focused on its use as solid fuel so far (Berge *et al*., 2015; Benavente *et al*., 2016; Owsianiak *et al*., 2016; Liu *et al*., 2017). These four studies showed how environmental performance of hydrochar used as solid fuel depends on the type of fuel that the hydrochar substitutes and on the incumbent waste management system that HTC replaces.

Global warming potentials (GWPs) are usually employed as indicators of climate change in LCA of products and systems (Laurent *et al*., 2012; Hauschild *et al*., 2013). In GWP, climate change impacts are expressed in terms of contribution of a greenhouse gas (GHG) to change in radiative forcing (not the actual warming) over a defined time horizon, typically 100 years (Forster *et al*., 2007). Global warming potential calculated for a 100-year time horizon (referred to as GWP100) is commonly used, standardized approach for assessing climate change impacts in LCA and carbon footprinting (e.g., ISO 14064, ISO 14067) (Laurent & Owsianiak, 2017). In addition to GWP100, the global temperature change potential at 100 years, GTP100, has been proposed (Shine *et al*., 2007; Levasseur *et al*., 2017). It uses as an indicator the global average temperature increase of the atmosphere at a future point in time (here, 100 years) that results from the emission (Shine *et al*., 2007). GTP100 is deemed as the most appropriate indicator to capture climate change impacts from gases with long residence times in the atmosphere, like CO_2. The third complementary indicator of climate change is the climate tipping potential (CTP), developed recently by Jørgensen *et al*. (2014a, 2015). The CTP expresses the contribution of a GHG emission to crossing a critical climatic target level (e.g., at 450 ppm CO_2 eq.) and is defined as climate impact relative to remaining capacity of the atmosphere for receiving GHG emissions without exceeding the atmospheric target level. Compared to GWP100 and GTP100, the CTP is the indicator with the shortest perspective as it addresses impacts occurring within decades, and gives more weight to short-lived GHGs like methane. Because of different perspectives that the three indicators offer, they are complementary to each other and are considered as different life cycle impact categories in LCA (Jørgensen *et al*., 2014a; Levasseur *et al*., 2017). Their use in LCA can potentially offer new insights into the climate change mitigation potential of hydrochars. This has, however, not been studied until now.

The aim of our study was therefore to evaluate the application of hydrochar to agricultural soils as a potential technology for carbon sequestration and temporary storage, using three indicators of climate change (namely GWP100, GTP100, and CTP), while taking into account uncertainties caused by yet incomplete knowledge about the influence of hydrochar on crop productivity, hydrochar stability in the soils, and emissions of GHGs from the soil as influenced by the hydrochar. Although climate change impacts are the main focus of this paper, we report full life cycle inventory and life cycle impact assessment results presenting impact scores (ISs) for 17 categories of environmental impacts. We thereby acknowledge that a broad spectrum of potential environmental problems, going beyond just climate change, is relevant for decision making about hydrochar use in agriculture.

Materials and methods

In the below, we present the study design and methods used to carry out literature review as basis for defining scenarios and model parameters in our LCA. Details of the LCA methodology are presented thereafter.

Study design

Figure 1 shows major methodological steps. As a starting point, we collected empirical data from a literature review to support defining relevant scenarios and model parameters for the LCA. We systematically reviewed effects of hydrochar on crop productivity, kinetics of evolution of CO_2 derived from mineralization of hydrochar carbon, and effect of hydrochar on priming of mineralization of native soil organic carbon, CH_4 fluxes, and N_2O fluxes from/into the soil. These reviewed data are used as input for performing inventory modeling and subsequent life cycle impact assessment including quantification of sensitivity and uncertainty. Life cycle impact assessment was carried out considering all relevant categories of environmental impact, including the three indicators of climate change: global warming, global temperature change, and climate tipping. The results were used to provide recommendations to decision makers about the use of hydrochar in agriculture as a soil conditioner, and recommendations for LCA practitioners and method developers about the application of the three different climate indicators in LCA.

Literature review and data treatment

A comprehensive review of Reza *et al*. (2014) was taken as starting point to identify papers which might contain relevant data on the effects of hydrochar on crop productivity and soil emissions. To complement their review, additional new data were retrieved from peer-reviewed studies available until

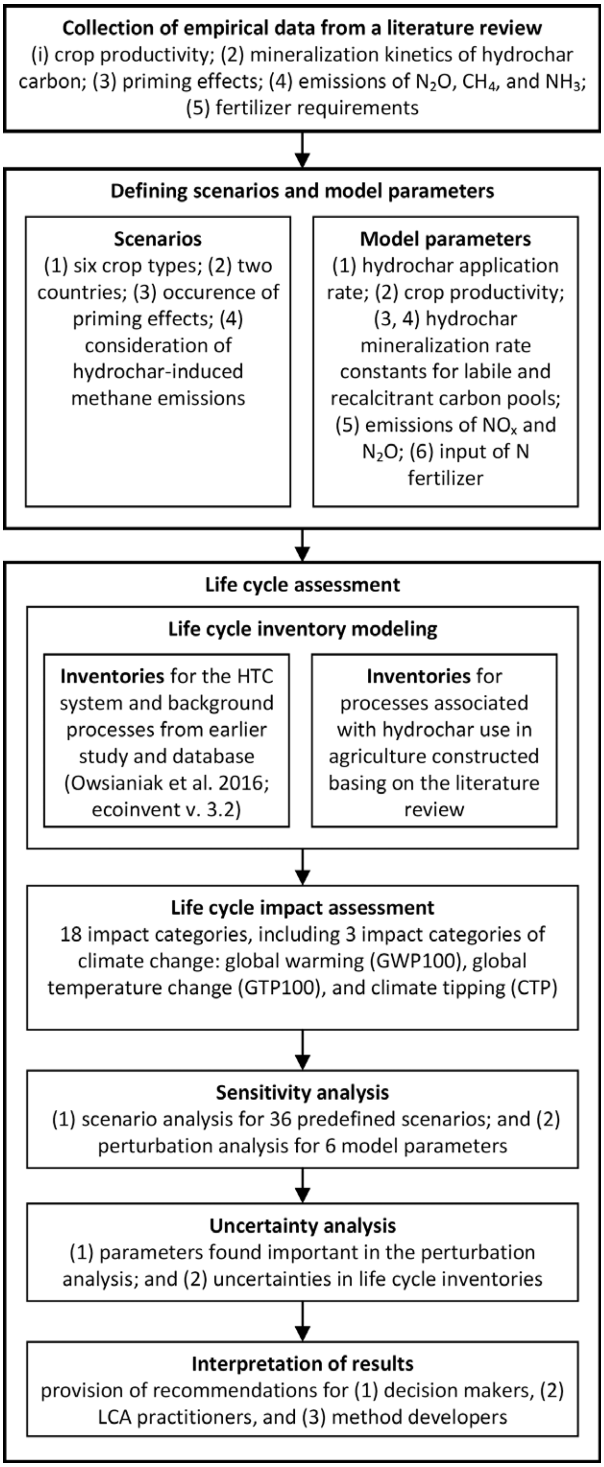

Fig. 1 Overview of major methodological steps in the study.

March 2017 identified through searching the ISI Web of Knowledge, version 5.7 (Thomson Reuters, New York, NY), using a combination of keywords: (i) soil and either (a) hydrochar or (b) hydrothermal. Citation lists of studies that contain potentially relevant data were then consulted to complement the search, and furthermore, all retrieved studies were screened in ISI to identify studies which cited them. These steps were iterated until no new studies were found. The data collected in the literature review were critically assessed and used in defining model parameters for modeling life cycle inventories. For this purpose, we quantified medians, geometric means, geometric standard deviations, and ranges of collected values. The collected data are documented and analyzed in detail in the SI, Appendix S1 (Tables S1-S5). An overview of criteria for inclusion of data into the study is presented below.

Crop productivity. Data on crop productivity were included if two criteria were met: (i) Experiments were performed with crops grown in soils (thus, excluding soil-less cultures); and (ii) hydrochar was the sole carbon source (thus excluding hydrochars mixed with raw feedstock or organic fertilizers like manure). To increase the number of data, we had to combine information from experiments performed either with or without addition of inorganic N, P, or K fertilizers. Furthermore, we included data from both pot and field experiments. Both plant biomass and grain yield were included as indicators of the effects of hydrochar on crop productivity (Busch *et al.*, 2012; Gajić & Koch, 2012; George *et al.*, 2012; Bargmann *et al.*, 2014a,b; Reibe *et al.*, 2014, 2015; Wagner & Kaupenjohann, 2014).

Mineralization kinetics of hydrochar carbon. Hydrochar contains carbon pools of different stability in soils and the mineralization of hydrochar carbon often follows biexponential decay kinetics (e.g., Bai *et al.*, 2013). Thus, data on the content of recalcitrant and labile carbon pools and the respective mineralization kinetic parameters were only included if derived from biexponential models. Furthermore, we combined data from studies which quantified amounts of CO_2 using one of the following methods: alkaline solutions used as CO_2 traps combined with titration (Gajić *et al.*, 2012; Qayyum *et al.*, 2012), methods using incubation vessels combined with gas chromatography (Dicke *et al.*, 2014; Schulze *et al.*, 2016), methods using an automated gas analysis systems (Lanza *et al.*, 2015), methods basing on measurements of isotope signature of CO_2 (d13C-CO_2) (Naisse *et al.*, 2014; Budai *et al.*, 2016), or methods measuring evolution of $^{13}CO_2$ from 13-C labeled carbon (Bai *et al.*, 2013). It was assumed that the evolved CO_2 is solely a result of hydrochar mineralization. We combined data irrespective of duration of the experiment.

Priming effects. To quantify mineralization of native soil organic carbon as influenced by hydrochar (e.g., either positive or negative priming) separately from mineralization of hydrochar C, we used methods based on measurement of isotopic composition of the evolved CO_2 (δ13C analysis (Malghani *et al.*, 2013; Bamminger *et al.*, 2014; Budai *et al.*, 2016). Only isotope-based methods allow distinguishing between CO_2 originating from hydrochar carbon and that from soil organic carbon. We excluded studies which report effects on priming using exogenous carbon sources, like glucose. Again, we combined data irrespective of duration of the experiment.

Emissions of nitrous oxide, methane, and ammonia. We combined data from all experiments reporting influence of hydrochar or emissions of N_2O, CH_4, and NH_3, irrespective of experimental techniques used for incubation and measurements, and again, irrespective of the duration of the experiment (Kammann *et al.*, 2012; Malghani *et al.*, 2013; Dicke *et al.*, 2014, 2015; Schimmelpfennig *et al.*, 2014; Subedi *et al.*, 2015).

Fertilizer requirements. Effects of hydrochar on improving soil fertility due to retaining nutrients (Libra *et al.*, 2011; Fang *et al.*, 2015) are difficult, if not impossible, to isolate from other hydrochar-induced effect, for example, the improved water retention properties. Thus, this parameter was not quantified, and our assumption about no change in fertilizer requirements was tested in sensitivity analysis.

Scenarios and model parameters

Scenarios. An overview of all 36 scenarios is presented in Table 1. Overall, we considered six different crops (barley, wheat, sugar beet, fava bean, onion, and lucerne) and two different countries (Spain and Germany). Except onion and fava bean, the crops chosen have been studied already in a hydrochar context and can be considered as potential crops for hydrochar applications. Spain and Germany were chosen because HTC plants are being developed in these countries (one of the first HTC plants has been erected in Spain) (Hitzl *et al.*, 2015), while European countries like Germany are important potential users of carbonaceous products in Europe (Ruysschaert *et al.*, 2016). Hydrochar-induced emissions of CH_4 and CO_2 into/from the soil (e.g., positive or negative priming of mineralization of native soil organic carbon and methane fluxes) are in current LCA practice not considered man-made and, thus not taken into account, but it could be argued that they are a result of human intervention and thus important for decision making about hydrochar use in agriculture. They were thus also considered in the scenario analysis. In all scenarios, we modeled hydrochar production from green waste for full commercial-scale HTC plant configuration with four reactors operating at capacity of 30 tonnes (dry weight) per day, as explained in Owsianiak *et al.* (2016). Green waste was chosen among other potential feedstock at it is relatively uncontaminated (it has heavy metal content comparable to that of food waste) and is relatively abundant in Europe (Karak *et al.*, 2012).

Model parameters. Based on findings from the literature review, we identified model parameters used to construct life cycle inventories (Table 2). Acknowledging that there is large variability and/or uncertainty associated with the estimations of these parameters, we defined baseline parameter values used as default in all scenarios listed in Table 1. We also defined perturbed parameter values representing lower and/or higher ranges of parameters as basis for perturbation analysis carried out to test the influence of a parameter value on the results, and as basis for comprehensive uncertainty analysis. Details of these analyses are presented together with description of LCA methodology in the sections Quantification of sensitivity and Quantification of uncertainty.

Life cycle assessment

The LCA was conducted in accordance with the requirements of the ISO14044 standard and the guidelines of the EU Commission's ILCD Handbook (ISO, 2006; EC-JRC, 2010).

Functional unit. The primary function of hydrochar in our context is to (temporarily) store carbon when added to agricultural soil. The functional unit, which ensures equivalence between all the compared systems, was therefore defined as "the average application and storage of 1 kg of biogenic HTC carbon to a temperate agricultural soil." This definition allows for a fair comparison between hydrochars with various stabilities in the soil (e.g., using parameter values reported in Table 2). A secondary function of hydrochar when applied to soil is its (potential) ability to support crop growth, and this property was also investigated by employing parameter values reported in Table 2.

Table 1 Overview of scenarios in the scenario analysis. Appendix S3 of the SI presents details of the accounting of hydrochar-induced CO_2 and CH_4 emissions

No.	Sensitivity parameter	Country	Crop type	Priming effect	Methane emissions
1	Baseline	ES	Barley	Not considered	Not considered
2–6	Crop type	ES	Wheat, sugar beet, fava bean, onion, lucerne	Not considered	Not considered
7–12	Geographic location of hydrochar production and use	DE	Barley, wheat, sugar beet, fava bean, onion, lucerne	Not considered	Not considered
13–24	Accounting for hydrochar-induced CO_2 emissions from mineralization of native soil organic carbon (positive priming effect)	DE, ES	Barley, wheat, sugar beet, fava bean, onion, Lucerne	Considered (137% of system without hydrochar)	Not considered
25–36	Accounting for hydrochar-induced methane emissions	DE, ES	Barley, wheat, sugar beet, fava bean, onion, lucerne	Not considered	Considered (418% of system without hydrochar)

Table 2 Model parameters for processes associated with hydrochar application to agricultural soils considered in the life cycle assessment (LCA) model. All values are based on measured values retrieved from literature review (see SI, Appendix S1). Parameters referred to as default apply to all scenarios listed in Table 1. Perturbation analysis was carried out to test the influence of a parameter value on the results for selected scenarios

Parameter	Parameter values		Unit	Description
	Default	Perturbation		
Application rate	5000	2500; 10000	kgC ha^{-1}	Application rate corresponds to that of 0.6% w/w content of hydrochar incorporated into 15 cm soil depth and is based on values used in pot experiments (Gajić & Koch, 2012). This value is in lower range of values usually tested experimentally in (where up to 10% w/w is used), and is expected to be within the range of values that would render field-scale application of hydrochar to soil practically feasible. We also considered two additional options: (i) 2500 kgC ha^{-1} and (ii) 10 000 kgC ha^{-1}. The former corresponds to nearly the smallest value tested experimentally (0.34% w/w), while the latter is expected to be below or close to the values that would be practically feasible in field-scale application (ca. 1% w/w)
Crop productivity	109	67; 178	% of system without hydrochar	Median productivity increase measured for all crops at relatively medium–low (≤4% w/w) application rates. We considered two alternative values: (i) 67% of system without hydrochar, being equal to the 5th percentile of values measured for crops at relatively medium–low (again, ≤4% w/w) application rates and (ii) 178% of system without hydrochar, being equal to the 95th percentile of values reported for all crops at medium–low application rates (again, at <4% w/w)
Mineralization rate constant for the labile pool*	0.081	0.012; 0.14	day^{-1}	Median mineralization rate constant for the labile carbon pool measured across hydrochars in soils. Perturbation included: (i) slow mineralization, with the values equal to the 5th percentile of values measured experimentally for hydrochars in soils (0.012 day^{-1}) and (ii) fast mineralization, with the values equal to 95th percentile of values measured experimentally for hydrochars (0.14 day^{-1})
Mineralization rate constant for the recalcitrant pool	0.0003	0.00014; 0.0014	day^{-1}	Median mineralization rate constant for the recalcitrant carbon pool measured across hydrochars in soils. Perturbation included: (i) slow mineralization, with the value equal to the 5th percentile of values measured experimentally for hydrochars in soils (0.00014 day^{-1}) and (ii) fast mineralization, with the value equal to the 95th percentile of values measured experimentally for hydrochars (0.0014 day^{-1})
Emissions of N_2O and NO_x	87	418	% of system without hydrochar	Average (geometric mean) value measured for hydrochars in soils. We also considered the 95th percentile of emissions measured experimentally equal to 418% of system without hydrochar. Emissions of N_2O were scaled to the N fertilizer input. Emissions of NO_x are linearly related to emissions of N_2O in ecoinvent processes and were thus scaled accordingly
Input of N fertilizer	100	50	% of system without hydrochar	No influence of hydrochar on fertilizer input was assumed in the baseline as number of studies on the effect of hydrochar on N fertilizer inputs is limited and findings rather inconclusive. We also considered one additional alternative: (i) 50% of system without hydrochar. Hydrochar produced from green waste materials contains relatively large amounts of N (1.7%, dry weight, ash-free) which might become a nutrient reservoir for plants (Reza *et al.*, 2014 and references therein). Thus, the 50% of system without hydrochar is deemed to be within range of realistic values. The ecoinvent processes for crop agriculture had to be modified to scale inputs (ammonium nitrate), emissions to air (dinitrogen monoxide, ammonia, nitrogen oxides), and emissions to water (nitrate) to different N fertilizer inputs

*Fraction of the labile pool was assumed equal to 0.034 kgC kgC^{-1}, which is an average (geometric mean) value measured across hydrochars. As the fraction of the labile pool was not found to vary largely across various hydrochars (variance equal to 0.006), this value was used consistently. Fraction of the recalcitrant pool was calculated as a difference between total carbon pool and the labile pool.

System boundaries of the assessment. Hydrochar application to soil is a prototype technology, while the HTC itself is a relatively immature option for biowaste treatment. Thus, the production and use of hydrochar as soil conditioner are not expected to have large structural changes on the market. Therefore, the current study is considered a microlevel decision support (type A) situation according to ILCD guidelines, and the assessment applies an attributional approach where average Spanish or German data and energy mixes are used. System boundaries specifying the processes included in the assessment are presented in Fig. 2. Details of the system boundaries with regard to the HTC and the hydrochar are described in Owsianiak *et al.* (2016). In addition to replaced conventional waste management system (composting or incineration, depending on the country), we also included HTC plant, production and postprocessing of the hydrochar and HTC process water, and transportation of the hydrochar. In cases of processes with recovery of commodities, system expansion was performed, where recycled steel substitutes the production of virgin steel and the HTC process water (concentrated at the HTC plant using reverse osmosis) substitutes the production of inorganic fertilizers. Likewise, impact offsets (also known as credits) are given to avoided agriculture, and to avoided conventional treatment of biowaste in accordance with the recommendations of the ILCD guidelines for this decision support type.

System modeling. The product systems were modeled in SimaPro, version 8.3.0.0 (PRé Consultants bv, the Netherlands). Data for foreground processes in the HTC system, like types of equipment and material and energy inputs for the plant, are based on primary data measured at a HTC plant at Ingelia S.L. (Valencia region, Spain). Data for generic processes, such as electricity production and waste management processes, are based on those available in the ecoinvent database, version 3.2 (Weidema *et al.*, 2013). Ecoinvent is currently one of the most comprehensive databases of life cycle inventories (i.e., aggregates of resource consumptions and pollutant emissions for specific processes taken in their life cycle perspective). Parameters and data underlying the modeling of HTC plant and post-treatment equipment are documented in Owsianiak *et al.* (2016) (see Table S3 in their study).

Impact assessment. Environmental ISs were calculated using the ILCD method for life cycle impact assessment (ILCD 2011 Midpoint+, version 1.05) (Hauschild *et al.*, 2013), as implemented in the LCA modeling software SimaPro, version 8.3.0.0 (PRé Consultants bv, the Netherlands). Apart from ionizing radiation impacts on ecosystems considered not sufficiently representative for this type of impact, all ILCD impact categories were considered: global warming using GWP100 of IPCC (2013), stratospheric ozone depletion, photochemical ozone formation, terrestrial acidification, terrestrial, freshwater and marine eutrophication, toxicity of released chemicals on freshwater ecosystems (termed “freshwater ecotoxicity” in the following) and on human health (termed “human toxicity,” differentiated between cancer and noncancer effects), particulate

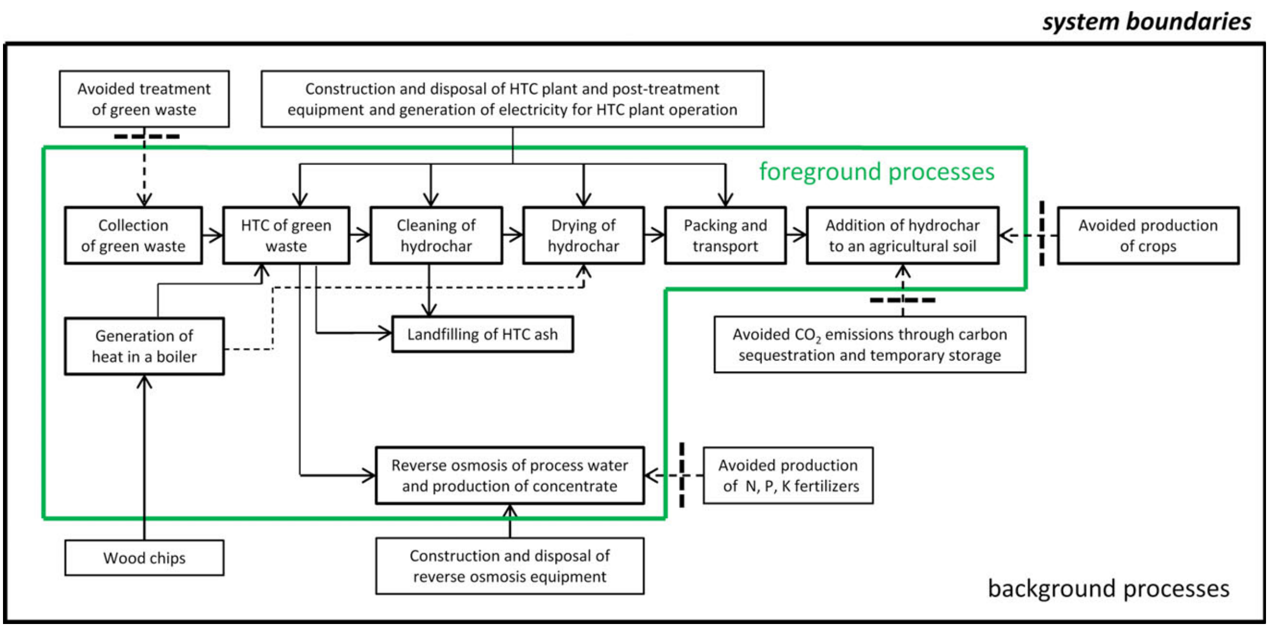

Fig. 2 System boundaries for hydrothermal carbonization (HTC) of biowaste with hydrochar application to an agricultural soil for carbon sequestration and temporary storage. The functional unit is “the average application and storage of 1 kg of biogenic HTC carbon to a temperate agricultural soil.” Foreground processes refer to those processes which can be structurally changed by the decision maker, like HTC and soil conditioning. They were constructed based on findings from literature review (for the soil conditioning) combined with unit processes from earlier study by Owsianiak *et al.* (2016). Processes in the background system can typically not be structurally changed by the decision maker and thus were modeled using generic processes from the ecoinvent database, version 3.2 (Weidema *et al.*, 2013).

matter formation, impacts of radioactive substance on human health (termed "ionizing radiation"), land use, water use, and mineral, metal, and fossil resource depletion (Hauschild *et al.*, 2013).

In addition to the use of GWP100, which is the default metric used in the ILCD life cycle impact assessment method, we use two complementary metrics, namely the global temperature change potential (GTP100) and the CTP. These indicators were chosen among other indicators as they (i) are relevant to hydrochar systems (due to specific kinetics of CO_2 emissions from the hydrochar); (ii) can be used by LCA practitioners with relatively small effort (Levasseur *et al.*, 2017); and (iii) represent wide range of different climate impacts. Other indicators could also be considered (e.g., Levasseur *et al.*, 2010), or existing indicators could be further improved (e.g., to account for dynamics of biomass regrowth; Guest *et al.*, 2013; Cherubini *et al.*, 2016), but their implementation was out of scope of this study. The three chosen indicators have different time perspectives and therefore represent different categories of impacts: from very short, nearly immediate perspective representing impacts stemming from the crossing of climatic tipping points (CTP), through longer but still relatively short/medium-term perspective for impacts stemming from increase in radiative forcing over the time horizon of 100 years (GWP100), to long-term impacts associated with increasing in mean surface temperature in 100 years (GTP100). Thus, the results are reported in parallel to the other 14 ILCD impact categories. Major features of the three climate change indicators are synthesized in Table 3. List of all LCIA methods with references is presented in the SI, Appendix S2.

Quantification of sensitivity

Scenario analysis. Sensitivity of the results to discrete parameters (e.g., crop type, geographic location, and accounting of CO_2 and methane emissions) presented in Table 1 was conducted by simply comparing ISs, without any internal normalization.

Perturbation analysis. For continuous parameters presented in Table 2, sensitivity of ISs was quantified using perturbation analysis, by varying an input parameter and observing the resulting change in IS relative to the result using the nonperturbed input parameter. Sensitivity of ISs was quantified by computing normalized sensitivity coefficients (eq 1), as done in e.g., Ryberg *et al.* (2015):

$$X_{\mathrm{IS},k} = \frac{\Delta \mathrm{IS}/\mathrm{IS}}{\Delta a_k/a_k} \quad (1)$$

where $X_{\mathrm{IS},k}$ is the normalized sensitivity coefficient of IS for perturbance of continuous parameter k, a_k is the kth parameter value, Δa_k is the perturbation of parameter a_k, IS is the calculated IS, and ΔIS is the change of the IS that resulted from the perturbation of parameter a_k. Note, that the Δa_k is chosen based on the realistic ranges of parameter values. A parameter is considered important if $X_{\mathrm{IS},k} \geq 0.5$, corresponding to a large sensitivity (Cohen *et al.*, 2013).

Quantification of uncertainty

We considered uncertainties in the parameters which were found important in the perturbation analysis (i.e., $X_{\mathrm{IS},k} \geq 0.5$), namely mineralization rate constants and crop yield. They were assigned geometric standard deviations based on the distribution of measured values retrieved in the literature review, following the method presented in Huijbregts *et al.* (2003) (SI, Appendix S4). Uncertainties in the life cycle inventories for the foreground processes (e.g., in material inputs or emissions) were estimated using the Pedigree matrix approach (Ciroth, 2013), as done in Owsianiak *et al.* (2016), whereas uncertainties in the background processes were based on geometric standard deviations already assigned to flows in the ecoinvent processes that were used to create the background system. Monte Carlo simulations (1000 iterations) were carried out for pairwise comparison between the baseline scenario and other scenarios listed in Table 1 while keeping track of the correlations between the two systems. Comparisons were considered statistically significant if at least 95% of all 1000 Monte Carlo runs were favorable for one scenario.

Results

Data collected from the literature review are reported in the SI, Appendix S1. Life cycle inventories are reported in Appendix S3. Below, we present an overview of life cycle impact assessment results across all scenarios showing results for selected scenarios and impact categories. Results for all scenarios and all impact categories are documented in the Appendix S5.

Table 4 shows results in category-specific units across all 17 impact categories computed for the scenarios of barley agriculture in either Spain or Germany (scenarios 1 and 7 in Table 1, respectively). Figure 3 shows results for the three indicators of climate change for twelve scenarios testing the influence of the type of crop in either Spain or Germany (scenarios 1–12). Overall, four main trends can be observed. First, using hydrochar in agriculture may bring environmental benefits, depending on the impact category. In the baseline scenario of barley agriculture in Spain, environmental benefits are seen in six of 17 impact categories, including climate change (GWP100) and climate tipping, but not global temperature change (GTP100). Second, ISs are generally higher for Germany as compared to Spain. For barley, statistically significant differences between Spain and Germany were found in nine impact categories, and Germany performed worse in all categories except freshwater eutrophication (see Tables S12, SI Appendix S5). Third, although differences in ISs between crops might appear relatively small, we found statistically significant differences in the majority of impact categories (Tables S13-S14 in the SI, Appendix S5). The fourth main observation is the important contribution from emissions of methane and

Table 3 Major features of the three indicators of climate change used in this study. List of all 17 LCIA indicators is presented in the SI, Appendix S2

Name and reference	Global warming potential (GWP) (Forster *et al*., 2007)	Global temperature change potential (GTP) (Shine *et al*., 2007)	Climate tipping potential (CTP) (Jørgensen *et al*., 2014a, 2015)
Abbreviation and unit of indicator	GWP (e.g., GWP_{100}) in kg CO_2 eq. kg^{-1}	GTP (e.g., GTP_{100}) in kg CO_2 eq. kg^{-1}	CTP (e.g., CTP_{RCP6}) in ppt_{rc} kg^{-1} (parts per trillion of remaining capacity of the atmosphere to take up emission)
Definition	"integrated radiative forcing of a gas between the time of emission and a chosen time horizon, relative to that of CO_2" (Levasseur *et al*., 2017)	"global average temperature increase of the atmosphere at a future point in time that results from the emission determined for a specific time horizon divided by the temperature increase caused by an equivalent amount of CO_2" (Levasseur *et al*., 2017)	"absolute impact from a marginal GHG emission based on its share of the total impact that can still take place before a predefined target level is reached" (Jørgensen *et al*., 2014a)
Cause–effect description and time horizon (as used in this study)	Cumulative radiative forcing over 100 years*	Instantaneous temperature at 100 years†	Cumulative impact of a GHG emission relative to the atmospheric capacity for taking up GHG emissions before reaching the target level as it depends on the choice of target level (e.g., 450 ppm eq) and the development in atmospheric GHG concentration (e.g., in representative concentration pathway RCP6 scenario)‡
Time perspective of impact assessment	Short/medium-term climate change ("rate of climate change, impacts related to the adaptation capacity of humans and ecosystems") (Levasseur *et al*., 2017)	Long-term climate change ("long-term temperature increase and related impacts on ecosystems and humans") (Levasseur *et al*., 2017)	Very short, nearly immediate perspective representing impacts stemming from the crossing of climatic tipping point at given target level (e.g., atmospheric CO_2 concentration of 450 ppm eq.)
Dealing with carbon sequestration and biogenic emissions of CO_2	CO_2 incorporated in biomass and biogenic emissions of CO_2 are assigned GWP100 equal to -1 and 1 kg CO_2 eq, respectively	CO_2 incorporated in biomass and biogenic emissions of CO_2 are disregarded as no recommendations are made about how to deal with biogenic CO_2 (Levasseur *et al*., 2017)	Uptake of CO_2 is treated as negative emissions for storage occurring before target time, but biogenic emissions of CO_2 are assigned CTP depending on the timing of emission before the target time
Dealing with temporary carbon storage and delayed emissions	Delayed CO_2 emissions are given credits following the assumption that storing 1 kg CO_2 eq. during 100 years compensates a 1 kg CO_2 eq emission	Disregards any benefits from temporary carbon storage and just uses GTP100 values applied to relevant GHG emissions irrespective of when they occur	Carbon sequestered from the atmosphere and later stored is given credits only when stored sufficiently long beyond target time
Substance coverage	Vast majority of relevant GHGs including chlorofluorocarbons, hydrofluorocarbons, perfluorocarbons, and sulfur hexafluoride	Major GHGs only, including CO_2, CH_4, N_2O, HCF-134a, CFC-11, PFC-14, and sulfur hexafluoride	Three major GHGs only: CO_2, CH_4, and N_2O

(continued)

Table 3 (continued)

Name and reference	Global warming potential (GWP) (Forster *et al.*, 2007)	Global temperature change potential (GTP) (Shine *et al.*, 2007)	Climate tipping potential (CTP) (Jørgensen *et al.*, 2014a, 2015)
Stakeholder acceptance and use	Widely accepted and used indicator, employed in LCA and carbon footprinting, although there can be differences in approaches to dealing with biogenic carbon and delayed emissions (Christensen *et al.*, 2009; Laurent & Owsianiak, 2017)	Recommended by IPCC and LCA community, although not widely used in LCA studies (Levasseur *et al.*, 2017)	Relatively new indicator that has not been widely used, except demonstration case studies (Jørgensen *et al.*, 2014b, 2015)

*GWPs for shorter or longer time horizons, like 20 and 500 years, can also be calculated.
†GTPs for shorter time horizons, like 20 or 50 years, can also be calculated.
‡CTPs for different concentration pathways like the mitigation scenario RCP3PD or high baseline scenario RCP8.5 can also be calculated.

Table 4 Characterized impacts and accompanying 95% probability ranges from Monte Carlo simulations, expressed in category-specific units for hydrochar used in barley agriculture in Spain (baseline, scenario 1 in Table 1) and in Germany (scenario 7 in Table 1). The probability ranges represent both parameter and inventory uncertainties, as explained in detail in the SI, Appendix S4. Results for other scenarios are tabulated in the SI, Appendix S5 (Tables S9 and S10). Statistical comparison between impact scores taking into account correlations is presented in Table S12 of the SI (Appendix S5)

		Impact score (95% probability range)	
Impact category	Unit	Scenario 1 (barley; Spain)	Scenario 7 (barley; Germany)
Climate change (GWP100)	kg CO_2 eq	−1 (−2.9 to 0.022)	0.54 (−1.1 to 1.3)
Climate change, long-term (GTP100)	kg CO_2 eq	0.07 (−0.53 to 0.53)	0.99 (0.1 to 1.4)
Climate tipping (RCP6 2017)	ppt_{rc}	−0.01 (−0.034 to 0.0044)	−0.0077 (−0.034 to 0.0013)
Ozone depletion	kg CFC-11 eq	1.0E-07 (8.4E-08 to 1.4E-07)	9.4E-07 (7.2E-07 to 1.3E-06)
Photochemical ozone formation	kg NMVOC eq	5.3E-04 (−1.1E-03 to 1.8E-03)	2.3E-03 (−8.8E-04 to 1.0E-02)
Acidification	molc H+ eq	−0.007 (−0.017 to −0.0025)	0.0064 (0.0022 to 0.015)
Terrestrial eutrophication	molc N eq	−0.046 (−0.089 to −0.026)	0.0054 (−0.043 to 0.036)
Freshwater eutrophication	kg P eq	0.0018 (0.0014 to 0.0025)	0.002 (−0.0019 to 0.0031)
Marine eutrophication	kg N eq	−0.0014 (−0.0031 to −0.00042)	0.00084 (−0.0039 to 0.0033)
Freshwater ecotoxicity	CTUe	13 (10 to 16)	19 (−60 to 57)
Human toxicity, cancer effects	CTUh	8.7E-08 (5.1E-08 to 1.2E-07)	2.1E-07 (2.5E-08 to 4.7E-07)
Human toxicity, noncancer effects	CTUh	2.5E-06 (1.6E-06 to 5.0E-06)	6.0E-06 (−4.6E-05 to 5.4E-05)
Particulate matter	kg PM2.5 eq	7.3E-04 (3.9E-04 to 1.1E-03)	1.3E-03 (7.9E-04 to 2.2E-03)
Ionizing radiation, human health	kBq U235 eq	0.13 (0.12 to 0.16)	0.39 (0.15 to 3)
Water resource depletion	m^3 water eq	−0.05 (−0.13 to −0.015)	0.2 (−1.1 to 0.59)
Land use	kg C deficit	2.3 (−64 to 47)	0.74 (−60 to 57)
Mineral, fossil, and renewable resource depletion	kg Sb eq	8.1E-05 (6.0E-05 to 1.3E-04)	8.0E-05 (5.1E-05 to 1.5E-04)

CO_2 from mineralization of native organic carbon if these GHGs are accounted for, with much larger contribution from methane than that of CO_2 (Fig. S3, SI Appendix S5).

Overall, our results suggest that (i) hydrochar production and use in agriculture can bring environmental benefits, depending on the country of hydrochar production and use, and category of impact considered; (ii) consideration on the influence of hydrochar on emissions of methane and CO_2 from mineralization of native soil organic carbon is important; and (iii) different indicators of climate change provide compounding insights with regard to climate change mitigation potential of hydrochars.

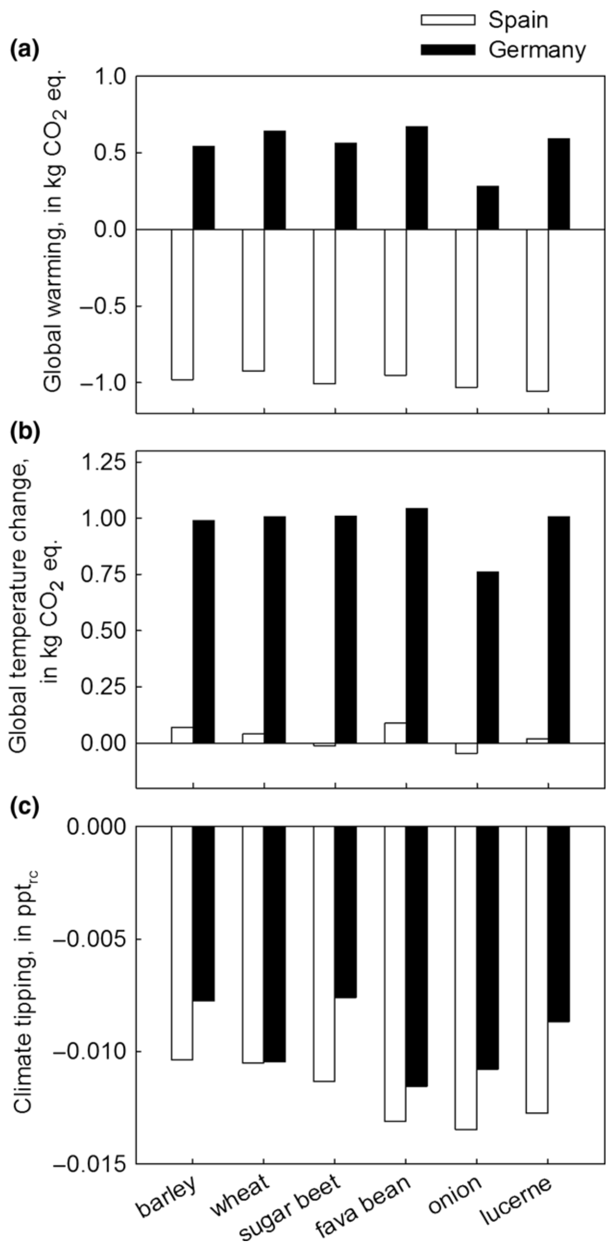

Fig. 3 Characterized impact scores in category-specific units for three climate change impact categories for hydrochar use in agriculture of either of six crops (barley, wheat, sugar beet, fava bean, onion, and lucerne) in either Spain or Germany (scenarios 1–12 in Table 1). The scores are for the functional unit defined as "the average application and storage of 1 kg of biogenic HTC carbon to a temperate agricultural soil." Absolute uncertainties are too large to be shown, but statistical comparison taking into account correlation between uncertainties revealed significant differences between countries and crops (see the SI, Appendix S5). Results for scenarios considering priming effects and increase in methane emissions (scenarios 13–24 in Table 1) are presented in the SI, Appendix S5 (Fig. S3).

Discussion

In the below, we explain results and evaluate of hydrochar as a potential carbon sequestration and storage technology. Implications for decision makers, LCA practitioners, and method developers are presented thereafter.

Insights from the three indicators of climate change

To explain differences between countries, a process contribution analysis, that is, identifying processes with the largest environmental burden, was conducted for the scenarios with barley agriculture in Spain and Germany (scenarios 1 and 7 in Table 1), complementing overall results presented in Table 4 and Fig. 3. As each indicator sheds light on a specific aspect of climate change impacts, results are interpreted per indicator. They are presented in Fig. 4.

Global warming. The process contribution analysis shows that climate change benefits from carbon sequestration and temporary storage, as quantified using the GWP100 approach, are the same in the two countries (Fig. 4a). Thus, the difference in ISs between Spain and Germany originates from different processes, in particular the waste management system that is replaced by HTC when green waste is treated hydrothermally. In Spain, HTC replaces composting with fertilizer recovery. Although there are some benefits from using compost as fertilizer (resulting in avoiding production of inorganic fertilizer), overall composting is not beneficial from the global warming perspective due to emissions of methane. Thus, replacing composting with HTC brings benefits to the hydrochar system. Figure 4a shows that these benefits are at least twice higher than benefits from carbon sequestration and temporary storage in hydrochar (ca. 0.3 kg CO_2 eq.), and are higher than burdens stemming from transportation of the biowaste (ca. 0.2 kg CO_2 eq), hydrochar production (ca. 0.3 kg CO_2 eq), and its application to soil by ploughing (ca. 0.05 kg CO_2 eq.). By contrast, replacing biowaste incineration with recovery of heat and electricity as primarily done in Germany does not bring climate benefits to the hydrochar system because the recovery of energy at the incinerator itself avoids emissions of fossil CO_2 which is an important contributor to global warming impacts from electricity and heat production in Germany. Although potential environmental benefits from carbon sequestration and temporary storage are not sufficient to outweigh climate burdens in this country, the reader should note that this rebound effect might not occur in countries with a cleaner grid mix.

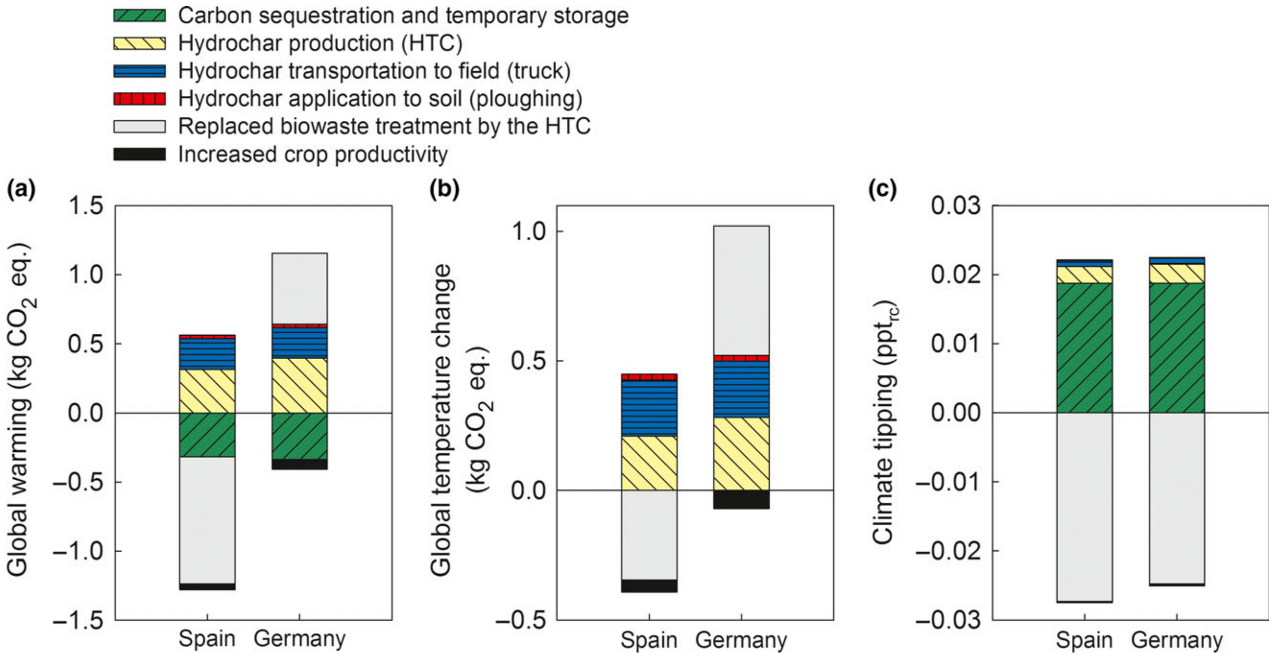

Fig. 4 Contribution of life cycle processes to total impacts from hydrochar use in agriculture of barley in either Spain (ES) or Germany (DE) (scenarios 1 and 7 in Table 1) presented for three climate change impact categories expressed in category-specific units (a: GWP100; b: GTP100; c: CTP).

Global temperature change. Assessment of climate change mitigation potential of hydrochar using GTP100 as an indicator generally shows no mitigation from using hydrochar in agriculture irrespective of the country, except for onion agriculture in Spain where negative scores are calculated (Fig. 4b). Process contribution analysis revealed that this is due to disregarding potential contribution from temporary carbon storage, currently not considered in the GTP100 approach. Potential benefits from replacing composting are smaller than in the GWP100 approach because GTP for methane is only 11 times larger than that of CO_2 (as compared to its GWP100 equal to 25 kg CO_2 eq.). Indeed, with its long-term perspective, GTP100 gives less weight to short-lived GHGs like methane and this influences the comparison in our case study. Omission of several chlorinated and/or fluorinated methanes and ethanes from our assessment due to missing GTP100 is not expected to influence our conclusions, because their contribution to climate change impacts in the GWP100 approach is very small (ca. 1%), and because they are not important GHGs in a biowaste treatment context.

Climate tipping. Climate tipping ISs are negative and equal to −0.01 and −0.0077 ppt_{rc} for Spain and Germany, respectively. Process contribution analysis revealed that these negative scores are, again, mainly due to replaced compositing of biowaste in Spain (Fig. 4c). These benefits are larger (relative to contribution from other processes) than in the GWP100 approach, however, because CTP of methane (for an emission occurring in 2017) is 85 times larger than that of CO_2. In the CTP approach, which has the shortest perspective of the three indicators, more weight is given to short-lived GHG like methane. There are also climate tipping benefits from avoided incineration calculated in Germany, which is mainly due to emissions of biogenic CO_2 when biowaste is incinerated. This is different from the GWP100 approach, where neutrality of biogenic carbon sequestered and immediately emitted is assumed, and is different from the GTP100 approach where biogenic CO_2 is not accounted for (as no recommendations exist yet about it; Levasseur *et al.*, 2017). Although sequestration and temporary storage of carbon are included in the CTP approach, climate tipping benefits are only when carbon is stored (sufficiently long), which is not the case for incineration of biowaste where no storage occurs. Temporary carbon storage does occur in case of hydrochar added to soil, but impacts stemming from hydrochar application to soils due to emissions of biogenic CO_2 as hydrochar mineralizes over time are larger than benefits from temporary carbon storage because large part of biogenic CO_2 will be emitted shortly before the climatic tipping point, where CTPs are the largest, resulting in climate impact rather than mitigation. Only a small part of hydrochar carbon is stored beyond the target time.

When does hydrochar bring environmental benefits?

Perturbation analysis for parameters presented in Table 2 identified inherent stability of the hydrochar in the soil (i.e., mineralization rate constant for the recalcitrant carbon pool) and crop yield as the most influential parameters on environmental performance of hydrochar (see Table S15 of the SI, Appendix S5). Uncertainty of these parameters was considered in our analysis. Yet, it could be argued that as experience with using hydrochar in agriculture grows and technology matures, both hydrochar stability and crop yield will be optimized. This will reduce the uncertainty while potentially increasing climate change mitigation potential of hydrochar. Increasing yields may also increase other life cycle impacts, beyond climate change. It is therefore useful to investigate whether there are conditions where hydrochar could bring enough benefits to outweigh all the burdens in locations where its use is not yet beneficial, like Germany, or to what extent it can increase climate benefits in locations like Spain.

An important parameter potentially contributing to climate change mitigation is the inherent stability of hydrochar in the soil. The influence of hydrochar stability on short/medium-term climate change (GWP100 approach) is illustrated in Fig. 5a. It shows how changes in contribution from temporary carbon storage to global warming (in terms of contribution to radiative forcing) over the time horizon of 100 years increase with an increase in hydrochar stability. When stability increased, corresponding to mineralization half-life of ca. 15 years (against ca. 5 years as default), benefits from temporary carbon storage are roughly tripled. This increase might seem significant, but it was not sufficient to outweigh global warming impacts stemming from other life cycle processes in Germany. The increase was not that important in Spain where climate benefits (GWP100) were always larger than burdens irrespective of the hydrochar stability in the soil (see Appendix S5, Table S17). This shows relatively small influence of hydrochar stability on short/medium-term climate change in these two countries. Figure 5b shows the influence of hydrochar stability on climate tipping impacts, which, contrary to the GWP100 approach, is the smallest for least-stable hydrochar while differences between the most stable hydrochar and the hydrochars with default stability (equal to median across measured values) are very small. This pattern was not unexpected considering timing of CO_2 emissions and magnitude of CTPs. The contribution to climate tipping impact is initially smaller for the most stable hydrochar, consistently with the GWP100 approach, because both emissions are relatively small and CTPs are relatively small. Yet, this contribution increases rapidly toward year 2032 because large part of emissions will occur shortly before year 2032, where CTPs are the largest. This explains why contribution to climate change mitigation is largest for the least-stable hydrochar despite the fact that most emissions occur shortly (within 2 years) after storage time. These impacts are overall larger than some benefits from temporary carbon storage beyond year 2032. Thus, although from the very short-term perspective the use of least-stable hydrochar in Germany appears most beneficial, it does come at the expense of increasing short/medium-term impact (see Appendix S5, Table S17). Least-stable hydrochar could be a climate sound option for use in Spain, as it reduces very short-term climate (tipping) impacts, without considerably worsening short/medium-term climate impacts (see Appendix S5, Table S16). Long-term perspective offered

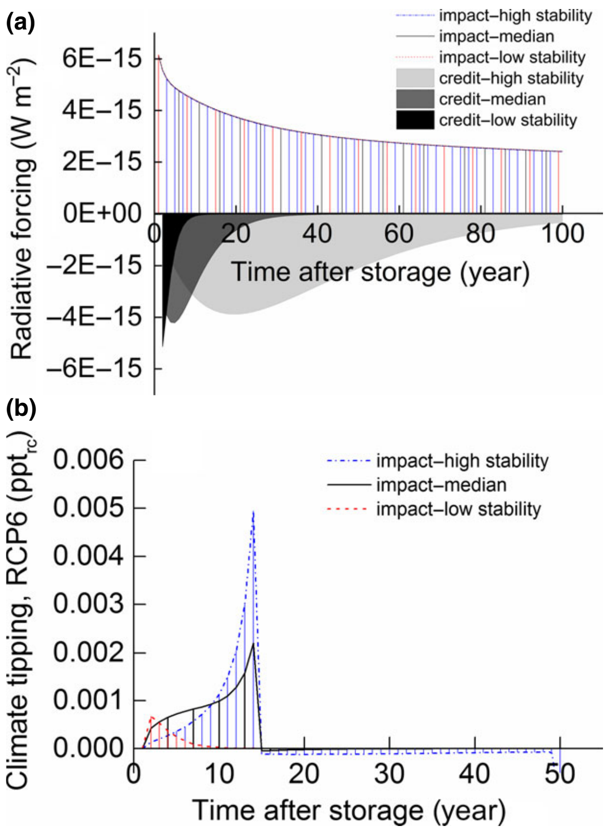

Fig. 5 Sensitivity of climate change impact scores to mineralization rate constant of the recalcitrant carbon pool in the hydrochar shown for the GWP100 approach expressed in terms of contribution to radiative forcing (a), and for climate tipping potential approach expressed in part per trillion of remaining capacity equivalents (b). The overall impacts represent area below the curves. Baseline value and perturbed values correspond to those presented in Table 2. The values just are for 1 kg of C stored and emitted as CO_2, disregarding other life cycle impacts. GTP indicator is not affected by stability of hydrochar and hence is not displayed.

by the GTP indicator in its current form is not affected by stability of hydrochar.

The second important parameter is the influence of hydrochar on crop productivity. When this parameter was equal to 109% of control (default value), the lowest ISs were consistently calculated for onion in both countries, while the highest were for fava bean in Spain and lucerne in Germany (Fig. 3). Although increasing yield to 178% of control, which is in higher range of measured values, increases climate benefits (GWP100) by a factor of 5–6 in Germany, these benefits do not outweigh burden as contribution of agriculture to total climate benefits is relatively small. Global warming impacts remain positive for all crops, except onion (see Table S19, Appendix S5), and the same trend was observed for the GTP100 approach. Relatively large impacts per tonne of onion produced, combined with relatively large inherent yields per hectare in Germany (40 tonnes ha^{-1}), result in overall large benefits when productivity increases. Overall, of all 17 categories of impact considered, the ISs are negative in 2 (lucerne), 3 (fava bean), 5 (barley, wheat, and sugar beet), and 11 (onion) impact categories when yield is equal to 178% of control in Germany (as compared with ISs being negative in 1 impact category in the scenario with the default value of 105%) (Tables S19). In Spain, where inherent yields are lower, increasing crop productivity in Spain to 178% of system without hydrochar addition would result in scores being negative in 8 (barley, wheat), 7 (sugar beet, fava bean, lucerne), and 10 (onion) impact categories (as compared to 5–7 impact categories when default value was used) (Table S19, Appendix S5).

Implications for implementation of the technology at field scale

This first life cycle-based evaluation of hydrochar as a potential carbon sequestration and temporary storage technology when used as soil conditioner highlights the key parameters which should be considered when making decisions about potential implementation of the technology at field scale.

We showed that although benefits from temporary storage of carbon are not negligible, they are relatively modest compared to impacts and benefits from replacing inefficient waste treatment options, like composting. Thus, climate change mitigation potential of hydrochars is mainly from replacing climate-inefficient waste management system. Carbon storage benefits from HTC can be either reinforced or counterbalanced by the type of waste management systems that it substitutes. Although the importance of a replaced waste management system has been shown in our earlier study on hydrochar used as solid fuel, here we show that it becomes even more important when hydrochar is used as soil conditioner because there are no benefits from substituting energy sources (Owsianiak *et al.*, 2016). As solid waste management systems are site- and country-specific, the overall performance of hydrochar systems will be case-specific. Thus, decision makers should carefully consider geographic location of hydrochar production and use, with focus on consideration of conventional biowaste management system within that location that the hydrothermal treatment replaces. Life cycle inventories described in the SI, Appendix S3, can be readily adapted to determine whether hydrochar production and use in agriculture in other geographic locations are valuable.

When the technology is implemented at field scale, focus will naturally be on ensuring that hydrochar increases crop productivity (e.g., through hydrochar washing to remove potentially toxic compounds). We showed that this parameter influences other types of impacts, not just climate change. Thus, all categories of impacts should be considered when supporting decisions about hydrochar use as soil conditioner. Decision makers should also note that although from the climate change perspective increasing yields might not always be sufficient to bring climate change mitigation, there will be environmental benefits in other impact categories, like terrestrial eutrophication and land use. This highlights the potential of hydrochar when its influence on crop productivity is optimized. Crop productivity will determine the cost and benefits of the technology and, ultimately, its practical implementation, and we showed that it is an important parameter to consider also from the environmental perspective.

Finally, although from a climate change mitigation perspective the choice of crop was found to be of relatively small importance, results presented in Appendix S5 of the SI and discussed in the previous paragraph clearly show that the response of hydrochar systems to this parameter is crop-specific. Hydrochar performs best for crops with inherently high yields per hectare (like onion), where benefits from increased productivity are the largest. It also performs well for crops which require relatively large inputs of fertilizer, like cereal crops, despite relatively low yields. By contrast, hydrochar is not expected to perform well for crops with low yields like fava bean or for crops which do not require fertilize inputs like lucerne. Our results for the scenario with lucerne also suggest that using hydrochar for just temporary carbon storage, for example, in areas where production of crops is not an important contributor to impact, like grass grown in grazing land, would not be a good idea from an life cycle perspective as benefits will not outweigh impacts even when high increases in productivity are foreseen.

Recommendations to life cycle assessment practitioners and method developers

Using three different indicators of climate change might seem challenging to LCA practitioners who need to calculate ISs and interpret results. We stress, however, that the three indicators are not alternatives to each other. On the contrary, they complement each other by offering different perspectives to quantifying climate change performance of a product or system. Our case study of hydrochar used as solid conditioner displayed this. Replacing composting with impacts driven by CH_4 emissions shows different trends between the short/medium-term perspective offered by the GWP100, where benefits due to CH_4 avoidance outweighing impacts from CO_2 emissions, and long-term perspective offered by the GTP100 where the opposite was the case. Replacing incineration shows generally no benefits in short/medium- and long-term perspectives because of the rebound effect on relatively dirty grid mix. Further, climate benefits from temporary storage of carbon also differ between indicators, indicating that there climate change mitigation is more consistently and thoroughly investigated when indicators offering different perspectives are employed. As the perspective can influence the assessment, we thus recommend practitioners quantifying climate change mitigation potential of products which release carbon temporarily, like hydrochars do, using different set of indicators chosen based on their relevance to the studied system. For hydrochar systems, we recommend LCA practitioners using global warming potentials (GWP100) and CTPs, both with credits given to temporary carbon storage, as these are particularly relevant to hydrochars which degrade relatively quickly in soils. The use of global temperature change potential indicators (GTP100) is also advocated as it focuses on long-term climate impacts, but it should be used and interpreted with caution when used for systems with temporary carbon as currently this indicator does not allow handling temporary carbon storage. For developers of impact assessment methods, the priority for method developers should be the harmonization of the three indicators used in this study in terms of substance coverage, and proposing recommendations about considering of temporary carbon storage in the GTP approach as recently was tested by others (Cherubini *et al.*, 2016).

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Supporting Information

Additional Supporting Information may be found online in the supporting information tab for this article:

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Appendix S6. References.