The crop residue conundrum: maintaining long-term soil organic carbon stocks while reinforcing the bioeconomy, compatible endeavors?

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The crop residue conundrum: maintaining long-term soil organic carbon stocks while reinforcing the bioeconomy, compatible endeavors?

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Abstract

Crop residues are key for supplying renewable carbon to the bioeconomy without interfering with food security. However, residue removal represents a challenge for soil organic carbon (SOC) stocks maintenance. This study demonstrates that the crop residues potential for the bioeconomy is spatially differentiated and depends on the conversion technology and the available recalcitrant carbon return to soils. We considered coproduct returns from five bioeconomy pathways: pyrolysis, gasification, hydrothermal liquefaction, anaerobic digestion, and lignocellulosic ethanol. Long-term SOC changes from these scenarios were compared against a reference where crop residues are unharvested. We developed an original framework by coupling a SOC model with a bioeconomy module, applicable to any site. The framework was tested and applied to the entire France, for 2020 – 2120, simulating more than 60,000 cropland units. It revealed an additional crop residue potential of 60 – 191 PJ (use-dependent) without SOC decreases, compared to the often used 31.5% removal limit.

Keywords: biochar; bioethanol molasses; digestate; hydrochar; recalcitrance; SOC modeling.

1. Introduction

Crop residues are a key feedstock to supply non-fossil carbon (C) to the future bioeconomy. In Europe alone, a theoretical potential of 3800 PJ y^{-1} [1] was estimated; equivalent to the gross annual electricity generation of France and Germany combined [2]. Crop residues include a variety of streams, such as (i) dry stalks and leaves of cereal and (ii) oilseed crops, and (iii) stems and leaves from tubers. These streams are leftover from harvest operations and thus not a primary economic product [3]. Current uses of crop residues include animal fodder and bedding, mushroom production, mulch to preserve soil moisture, among others [1,6].

When left unharvested, crop residues can contribute to soil organic carbon (SOC) and play a key role in the long-term quality, nutrient balance, and agronomic functions of soils. Increasing removal rates reduce the soil organic matter inputs, which creates a trade-off between the crop residue use for the bioeconomy and SOC stocks maintenance [7,8].

Various studies suggested limiting the removal to rates between 15% and 60% of the theoretical harvesting potential (depending on the crop type) due to technical and environmental constraints [6,9–12]. These restrictions significantly reduce the supply of renewable carbon from crop residues to the bioeconomy.

Bioeconomy processes convert biomass into a main product, while the more degradation-resistant fraction remains a coproduct. Coproducts can be applied to the soils as exogenous organic matter (EOM) to maintain or improve the SOC stocks [13]. EOM is a heterogeneous material and can be composed of recalcitrant and labile fractions. The labile fractions tend to be mineralized fast (i.e., as CO_2 emissions) after the first couple of years following soil application, while the recalcitrant fractions exhibit longer mean residence times (MRT; [14]), promoting SOC storage [14,15]. SOC stock evolution depends on the applied coproduct as well as the site-specific conditions and cropping systems (i.e., a combination of soil properties, climate, crop rotations, and other management practices). Spatially explicit considerations are thus needed in order to address the conundrum between long-term SOC storage and the supply of a renewable C feedstock to the bioeconomy.

Some soil C models can simulate long-term SOC dynamics considering different cropping systems, soil properties, and climates [16]. Organic matter decomposition involves complex processes influenced by the biomass characteristics, eventual stabilization treatment and/or recalcitrance degree, pedoclimatic conditions, and interactions with the soil microbiota, among others.

An accurate prediction of the coproducts' carbon persistence in soils is therefore challenging [17,18]. Some soil models have been adapted, or parameters have been proposed to simulate the return of bioeconomy coproducts into soils. This includes, for instance, RothC [19,20], Century [21], APSIM [22], and EPIC [23] for biochar; CTOOL [24], AMG [25], CANDY [26], and RothC [18] for digestate. RothC [18] has also been adapted to consider bioethanol coproducts, such as the non-fermentable residue. BioEsoil, a RothC-based tool, evaluates the effect of residues from bioenergy processes (i.e. gasification and incineration) on soil organic matter [27]. However, these studies have been site- and coproduct-specific, limited to very specific simulation parcels due to the scarcity of data and modeling issues to cover high spatial resolutions and temporal scales. To date, only a few studies have used a soil model that includes different EOM inputs coupled with large-scale spatial information, as in Mondini et al. [28], where eight types of EOMs were simulated at the scale of entire Italy.

The effect of crop residues harvesting and their use in the bioeconomy on long-term SOC stocks has been explored in Hansen et al. [24], where the authors found that for Danish soils, the residues that can be harvested for pathways involving no C return is 26% of what can be harvested on average, if residues are used for biogas, with 100% digestate return to soils. Yet, Hansen et al. [24] assessed only one bioeconomy conversion pathway and used a rather coarse spatial representation of Danish croplands limited to two types of crop rotations and three types of soils. Similarly, Woolf and Lehmann [20] predicted that applying biochar to soils could increase SOC stocks by 30–60% in 100 years while removing 50% of crop residues for bioenergy in three specific locations in Colombia, Kenya, and the USA.

To our knowledge, no study has addressed the effect on SOC stocks from crop residue removal and C return to the soil from various bioeconomy conversion pathways. In this work, we challenge the idea that the biomass potential from crop residues must be limited by a given removal rate to maintain organic carbon in arable soils. Instead, we propose that such potential is deeply intertwined with the use of the residual biomass within the bioeconomy. This is based on the rationale that many technologies involve a potential carbon return to soils as a coproduct more recalcitrant to degradation than the original raw biomass.

Thus, this study aims to (i) further understand the cause-effect link between the "C-neutral harvest" of crop residues (defined below) and their usage within the bioeconomy, and (ii) address how this differs among the major existing bioeconomy pathways where a C return to the soil is possible. To this end, we modeled, as an illustrative case study, the SOC evolution of all arable topsoils (0–30 cm) in France, with and without crop residues harvest for different bioeconomy pathways. The term "C-neutral harvest" is herein employed to designate situations where the long-term (here defined as 100 years) SOC stocks of a given bioeconomy management do not decrease, in comparison to a reference situation where crop residues are incorporated into soils. It encompasses a similar vision to what previous studies referred to as "sustainable harvest" [6] but is more explicit on what it covers (quantification of SOC stocks only) and what it disregards (e.g., other aspects of long-term sustainability such as biodiversity or soil fertility).

1. Methods

We propose a modeling framework to assess the long-term SOC stock effects of crop residues usage for different bioeconomy technologies, considering different management practices on arable topsoils. It quantifies the amount of harvestable crop residues that can be removed from fields for bioeconomy, while an alternative coproduct is returned to meet the condition of maintaining or even increasing SOC stock levels, as compared to a reference situation where residues are left on the field. The bioeconomy scenarios considered here include pyrolysis, gasification, hydrothermal liquefaction (HTL), anaerobic digestion (AD), and lignocellulosic bioethanol (2GEtOH) production.

The framework is based on spatially explicit high-resolution data on climate, soil, and agricultural practices of specific units of metropolitan France and a SOC model that simulates the SOC stock changes in cropping systems receiving coproduct inputs with specific recalcitrance properties. The temporal scope is 100 years, over the years 2020-2120.

The state of C in arable soils, agricultural practices, soil, and climate at the beginning of the time scope is here referred to as the initial condition. Two developments over the time scope are considered: (i) a business-as-usual (BAU) scenario reflecting current practices where part of the harvestable crop residues (ca. 46%; details in SI1.2) are already being exported for livestock (as bedding and fodder) and the rest is left on fields, and (ii) a bioeconomy scenario which is similar to the BAU, except that the share of straw left on fields is harvested, used in the bioeconomy (five distinct bioeconomy scenarios being independently considered), and partly returned to fields as a bioeconomy coproduct. The BAU scenario is thus a measure-stick against which the bioeconomy scenarios are contrasted. For these six future developments (i.e., BAU and the five bioeconomy scenarios), the long-term changes in SOC stocks are determined as further described in section 2.3.

The soil physicochemical properties and the future meteorological variables assumed (further detailed in sections 2.2 and 2.3) are the same for the BAU and bioeconomy scenarios. Similarly, the same rotations, farming management practices (section 2.2), and crop yields (SI1.2) are repeated cycle after cycle. The impact on crop yields, from changing the raw biomass to stabilized EOM, was excluded to emphasize the effect of crop residue removal alone [29]. The technical harvestable rate and amount of crop residues already used for livestock are crop-dependent (Table SI1.1) and are assumed to remain constant during the modeling timeframe.

The structure of the framework consists of four main modules (Fig. 1), further detailed in the subsequent subsections: description of the simulation units (Module 1), definition of the bioeconomy scenarios, carbon conversion from residues to coproducts, and recalcitrance of coproducts (Module 2), modeling SOC stock changes and collecting related input data (Module 3), and sensitivity analysis (Module 4). The process was automated using R [30]; the scripts and data used are available in Andrade et al. [31].

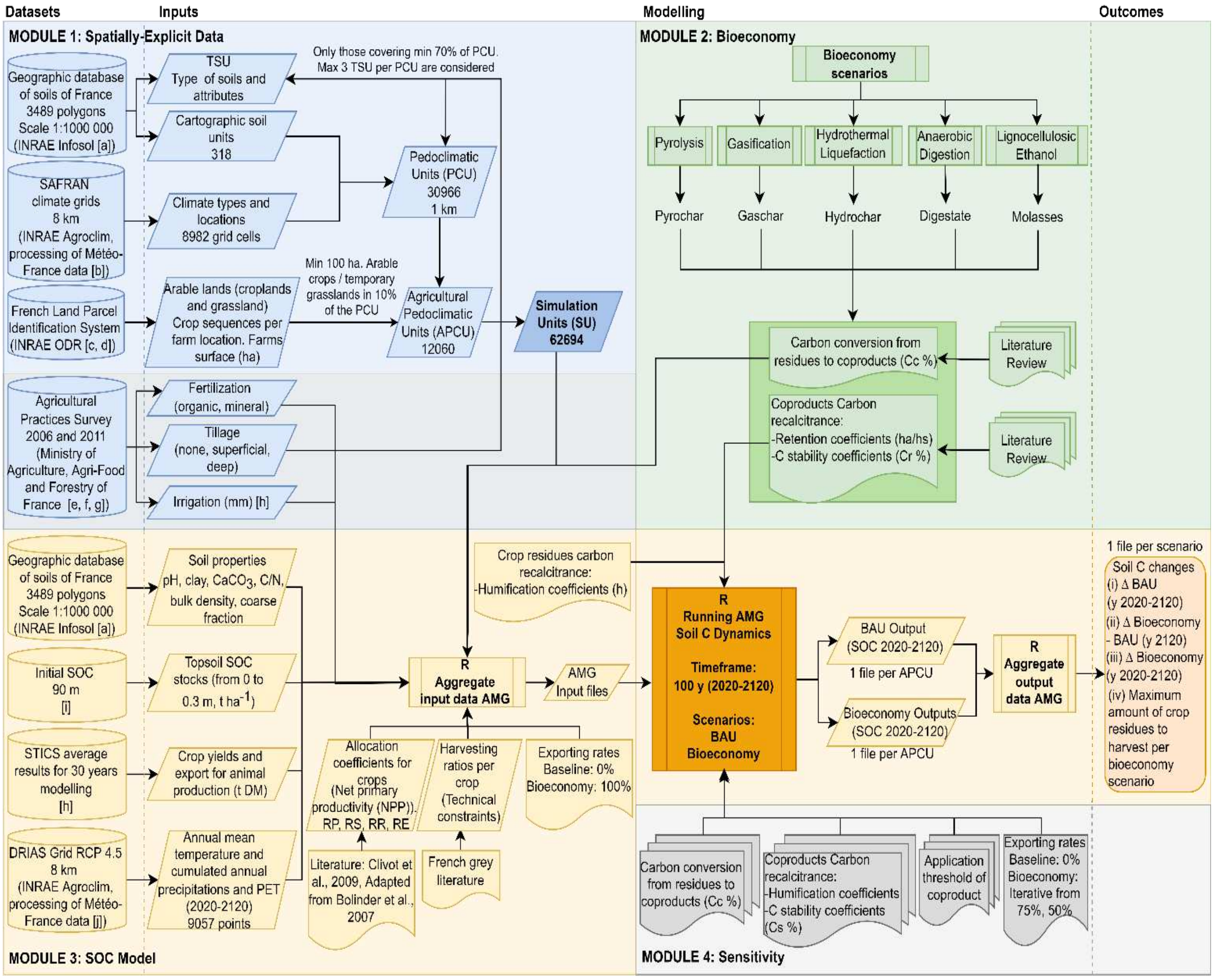

Fig. 1 Spatially explicit modeling framework, as applied to France, to quantify the long-term SOC difference between crop residues left on land and their harvest for five distinct bioeconomy pathways, with return of the coproduct. [a] [32], [b] Based on Durand et al.[33] after adaptation of Launay et al.[34], [c] [35], [d] [36], [e] [37], [f] [38], [g] [39], [h] [34], [i] [40], [j] CNRM-CERFACS-CM5--CNRM/ALADIN 63. Model GCM / RCM – correction ADAMONT. Institution : Méteo-France/CNRM, [41]. PET : Potential Evapotranspiration, RP: Relative Carbon allocation coefficient for the agricultural product, RR: Relative Carbon allocation coefficient for roots, RS: Relative Carbon allocation coefficient for straw or any post-harvest residue, RE: Relative Carbon allocation coefficient for extra root material, BAU: Business-as-Usual, SOC: Soil organic carbon, STICS: Soil-crop model used in Launay et al.[34], DRIAS: Spatially explicit database for France projections of climate scenarios, SAFRAN: Spatially explicit database for France climate. Figure legend: Cylinders: database, parallelogram: data input, rectangle: process, rectangle with inner bars: process containing more processes, curved bottom rectangle: manually input data sets, rounded rectangle: output.

2.1 Module 1: Spatially explicit data

The aim of Module 1 is to define representative simulation units (SU) for the studied case, reflecting the variety of soils, climates, crop rotations, and farming practices in a spatially explicit manner.

Module 1 is entirely building upon the study of Launay et al. [34], launched within the frame of the French efforts within the international 4p1000 initiative [42], acknowledged as the most comprehensive and updated spatially explicit representation of cropping systems in France. Launay et al. [34] defined a set of fundamental concepts briefly described as follows.

Pedoclimatic units (PCU) are defined as a unique combination of soil properties (coarse fraction, clay content, pH, etc.) and meteorological variables (temperature, precipitations, and potential evapotranspiration) (French climate and soil types are specified in SI1.1). When found on arable lands, these are referred to as agricultural PCU (APCU). French APCUs combine soil mapping units (1:1000,000; [32]) and the French SAFRAN climate grids (8x8 km; [33]) with identified crop rotations per PCU retrieved from the French Land Parcel Identification System [36,43]. A total of 12,060 APCUs with more than 100 ha of agricultural area, where at least 10% of it has arable crops and/or temporary grasslands, were identified. The selection represents 84% of the French cropland.

The crop rotations selected in Launay et al. [34] include 12 different crops, temporary grasslands, and cover crops (detailed in SI1.2). Winter wheat is the most representative crop, providing 65% of the available residual biomass (dry matter). Crop rotations cover 4.79 Mha and were judged to be a fair representation of the 18.35 Mha of French arable crops and temporary grasslands in 2006-2012. Farming practices —involving organic fertilization, cover crops, irrigation, tillage, and current use of crop residues— were determined from a survey conducted by the Ministry of Agriculture, Agri-Food, and Forestry over the period 2006-2011 [39,44].

The combination of APCUs, crop rotations, and farming practices yielded 62,694 simulation units (SI1.1). Further details on the crop rotation and yields are presented in SI1.2.

2.2 Module 2: Bioeconomy scenarios

This module describes the five bioeconomy scenarios. All the bioeconomy scenarios involve two key parameters to answer the research questions of this study, namely (i) the amount of C from the harvested crop residues (of a given SU; Fig. 1) that will end up in the coproducts returned to fields and (ii) the C recalcitrance to degradation of this coproduct. The former is hereafter referred to as carbon conversion (Cc) and the latter as carbon recalcitrance (Cr). The recalcitrance represents the most stable biochemical fraction of organic products. Here, Cr is the fraction of the coproduct that cannot be readily mineralized, and which decomposes slower than the more labile fraction of the organic coproduct. The labile fraction is assumed to be entirely processed by soil microorganisms within about one year. The Cc and Cr of a coproduct depend on the feedstock and process conditions.

The conversion pathways studied herein can be grouped as thermochemical (pyrolysis, gasification, and HTL) and biochemical technologies (AD and 2GEtOH production).

In this work, we only considered the return of the char produced in each thermochemical technology, and other coproducts generated (e.g., gas, tar, ashes) are excluded. To avoid confusion between the coproducts assessed in each scenario, we refer to pyrolysis, gasification, and HTL char as pyrochar, gaschar, and hydrochar, respectively. For the biochemical pathways, we consider only digestate (from AD) and molasses (from 2GEtOH production) as EOMs.

The Cr and Cc considered herein for these coproducts stem from a comprehensive compilation and data reconciliation of over 600 records from laboratory assays, field trials, and modeling experiments involving a wide variety of feedstock, including crop residues, as detailed in Andrade et al. [50]. To the extent possible, the Cc and Cr values used herein were derived from studies involving straw-like feedstock. Table 1 summarizes the Cc and Cr considered for each scenario. The full bioeconomy conversion pathways are further described in SI1.3.

Table 1. Overview of the bioeconomy scenarios considered in the study, and implications in terms of the Cc (Carbon conversion) and Cr (Carbon recalcitrance)

parameters. MRT: Mean Residence time, DM: dry matter, n/a: not applicable. In bold: intended (main) product of the conversion pathway.

Scenario	**Process Conditions**	**Coproduct returned to soil**	**Other products generated**[a]	**Cc**[b] **%**	**Cr**[b] **%**	**MRT**[c] **Years**	**Key process reference**[h]
BAU	Crops residues left on soil	None	None	n/a	n/a	n/a	[34]
Pyrolysis[d]	350 – 700 °C, from seconds to 2h, typically fed with a biomass DM>90%	Pyrochar[e]	**Bio-oil**, non-condensable gases	44 [34 – 54]	95 [90 – 99]	> 100	[14,51,52]
Gasification	600-1200°C dry gasification. 300-550°C hydrothermal gasification, typically fed with a DM>90%	Gaschar	**Syngas**, tar, ashes	20 [14 – 25]	95 [90 – 99]	> 100	[53–55]
Hydrothermal Liquefaction[f]	180-400°C, use of K_2CO_3 catalyst to enhance bio-oil production; typically fed with a DM<20%	Hydrochar	**Bio-oil**, non-condensable gases	31 [12 – 45]	83 [80 – 96]	< 26	[56,57]
Anaerobic[g] Digestion	Mesophilic conditions (30-50°C). 1-3 months. Typically, wet digestion, with DM in the digester<35%	Digestate	**Biogas**	33 [30 – 40]	68 [58 – 77]	< 26	[58–60]
Lignocellulosic ethanol	Pretreatment, acid and enzymatic hydrolysis, fermentation *S. cerevisiae, purification by* distillation. The effluent is separated	Molasses	**Ethanol**, solid fraction	24 [18 – 30]	45 [28 – 60]	< 26	[5,61,62]

into a solid fraction
and liquid molasses

[a] The main product considered to drive the investment in this bioeconomy scenario, under the specified conditions, is indicated in bold; [b] Cc: C fraction of initial crop residue transferred to the co-product returned to fields, Cr: C fraction of the co-product allocated to the stable biochemical fraction. The values presented herein are averages from Andrade et al. [50], based on a compilation of 124, 33, 97, 99, and 51 records, for pyrochar, gaschar, hydrochar, digestate, and molasses, respectively. Ranges in brackets represent quartiles 1 and 3; [c] The MRT allows to define the SOC fraction of the soil model to allocate the Cr fraction. The soil model used in this study considers that EOMs with MRT of the Cr fraction higher than the modeling timeframe are inert, thus any coproduct with an MRT longer than 100 years is virtually inert. EOMs with Cr fractions exhibiting MRT of 7 – 26 years are considered to be slowly mineralized [63] in the soil model (see section 2.3). [d] Pyrolysis can be classified as fast (300 – 500°C, seconds of retention time) or slow (500-700°C, minutes to hours). Slow conditions tend to favor the production of biochar, whereas the fast process is optimal for bio-oil production. From an economic standpoint, the pyrolysis scenario in this study aims to maximize the bio-oil yields, thus the process conditions of the studies included are those of a fast process, when possible [51]. [e] Also commonly referred to as biochar. [f] The use of catalysts, specially K_2CO_3 accelerates the water gas shift reaction in low temperatures HTL processes, which yields higher rates of bio-oil (targeted product) than hydrochar. The use of catalysts tends to be more common [64], therefore, the Cc and Cr values stem from such process conditions. [g] Some simulation units involve the use of manure as organic fertilizer. For these, we did not consider this manure to be digested, to keep the focus on the impacts from crop residues. Cc accounts only for the C from crop residues transferred to the digestate. [h] Only key references mentioned, the full compilation of reviewed studies is presented in Andrade et al. [50].

2.3 Module 3: SOC Model

Module 3 describes the SOC model used and the adaptations considered in this study, as well as how the bioeconomy scenarios have been compared to the BAU scenario.

2.3.1AMG model: Overview

For both the BAU and bioeconomy scenarios, the evolution of topsoil organic C stocks (0-30 cm) was simulated with the AMGv2 SOC model, detailed in Clivot et al. [63]. AMG is a French SOC model, first described in Andriulo et al. [65], which simulates the carbon dynamics of agricultural topsoils at an annual timestep. The model successfully predicted the changes in SOC stocks of various cropping systems under different pedoclimatic conditions in France and Europe [63,66,67] and has notably been calibrated for 26 EOM types [25]. AMG splits the organic matter (OM) into three different pools (shown as boxes in Fig. 2). The model structure, including the pools and parameters to allocate carbon in the model are further described in SI1.4.

2.3.2Model input data

AMG minimum input data comprises crop rotations, climate, soil physicochemical properties, initial SOC stocks, and farming practices (including the maximal soil tillage depth, irrigation water amounts, EOM inputs, and crop residue management). Crop rotation information includes annual yield, moisture content of the harvested product, harvest indexes (HI), and C allocation coefficients determining the proportion of C in the harvested product (RP), above-ground residues (RS), root C (RR), and extra-root C (RE) (Fig. 1). It also includes the fraction of residues that can be technically harvested, per crop type.

HI and allocation coefficients were used as set in the method proposed for calculating C inputs for AMGv2 [63], adapted from Bolinder et al. [68]. The technically harvestable fraction for each crop was also taken as defined in the proposed method (further details in Table SI1.1) and varies from 55% - 91%. Meteorological data comprises the mean annual air temperature and the annual water balance, the latter being determined as the difference between the water inputs (accumulated precipitations and irrigation) and potential evapotranspiration. In this study, the spatially explicit meteorological data were retrieved, for years 2020 to 2100, from SICLIMA (last updated May 2013 [69]), for the RCP4.5 climate trajectory (Representative Concentration Pathway [70]), downscaled by the model CNRM-CERFACS-CM5/CNRM-ALADIN63. These projections were not available beyond 2100. For the period from 2101 to 2120, average values from the last decade (i.e., from 2091 to 2100) were used.

Soil-related data include initial SOC stocks, pH, bulk density, coarse fraction, clay content, C:N ratio, and CaCO3 content. Initial SOC stocks were retrieved from Mulder et al. [40] and used as processed in Launay et al. [34] to correspond to the APCU resolution, while the other soil parameters were retrieved from Jamagne et al. [32]. AMG also requires information regarding farming practices, as detailed in Module 1.

Default C retention coefficients (*h*) (FOM-dependent) given in AMG for crop residues and non-coproduct EOMs (e.g., animal manure) [25,63] were used, while for the bioeconomy coproducts, these were determined individually as further detailed. The actual mineralization rate (*k)* of the active SOC pool, which depends on environmental response functions, is calculated for each year and each situation as defined in Clivot et al. [63].

2.3.3AMG adapted for bioeconomy processes

We adapted the calculation method for C inputs in the AMG model to include the Cc and Cr values of pyrochar, gaschar, hydrochar, digestate, and lignocellulosic ethanol molasses. The adapted version of AMG allows determining the SOC evolution of the different bioeconomy scenarios by deriving retention coefficients from Cr values. The C input from the coproducts is determined using the initial C in the crop residues and the Cc coefficient.

We grouped the Cr values per coproduct as highly recalcitrant (pyrochar and gaschar) and less recalcitrant (hydrochar, digestate, and molasses), to define the C retention in the soil associated with each coproduct. The recalcitrance and the MRT values in Table 1 were used to set the h coefficient per coproduct and allocate the C among the soil active and stable SOC pools (C_A or C_S), respectively. Two retention coefficients were defined to differentiate between the fraction integrating the active pool (h_a) and the stable pool (h_s).

For the highly recalcitrant coproducts, with MRTs longer than the modeling timeframe, the Cr fraction (95%; Table 1) was considered virtually inert and was directly allocated into the stable pool as h_s (Fig 2b), while the labile fraction was assumed to be entirely lost as CO_2 and no h_a was considered. For the less recalcitrant coproducts hydrochar, digestate, and bioethanol molasses, we assumed that the labile fraction (1-Cr; 17%, 32%, and 36%, respectively) was mineralized in the first year and the remaining recalcitrant fraction corresponded to h_a (here equivalent to Cr) and was fully allocated to the active pool (Fig. 2a). The derived h_a coefficient for the digestate (0.68) is close to the value of 0.65 proposed in Levavasseur et al. [25]. No reference allowing for similar comparison was found for the other studied coproducts. The adaptation of AMG for the bioeconomy is further detailed in SI1.4.

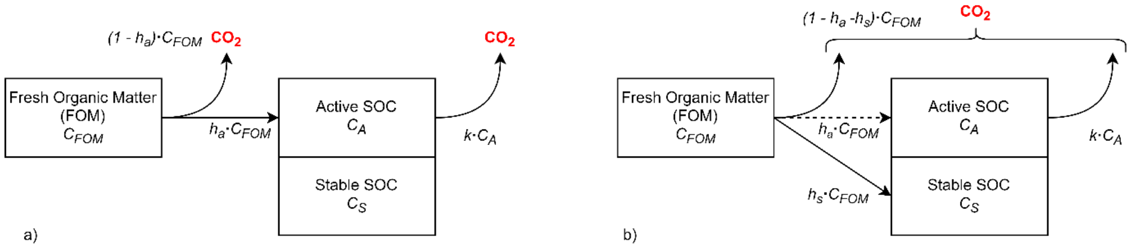

Fig. 2 AMGv2 configuration implemented in this study (adapted from Clivot et al., [63]): a) AMG v2 (for all streams but pyro- and gaschar), and b) adaptation of AMGv2 (for pyro- and gaschar). FOM: fresh organic matter, C_{FOM}: carbon in the fresh organic matter, $h_a.C_{FOM}$: fraction of C_{FOM} allocated to the active pool C_A, $h_s.C_{FOM}$: fraction of C_{FOM} allocated into the stable pool C_S, h_a: retention coefficient integrating a fraction of FOM into the active pool, h_s: retention coefficient integrating a fraction of FOM into the stable pool, k: mineralization rate constant, dotted line: FOM fraction allocated to the active SOC pool (see section 2.5).

2.3.4 AMG output analysis

The 100-year SOC changes were determined for each bioeconomy scenario and compared to the BAU. The SOC change per scenario (from 2020 to 2120, %), difference in SOC change between scenarios (bioeconomy vs BAU, %), and total net SOC change between bioeconomy and BAU (Mt C) were determined using equations S1, S2, and S3, as detailed in the SI1.4.

2.4 Module 4: Sensitivity Analysis

SOC stock changes are influenced by the characteristics and amount of the carbon inputs [71]. We performed a sensitivity analysis (SA) on the key parameters governing the amount of C returned to the soil contributing to SOC, namely Cc and Cr. As shown in Table 1, both Cc and Cr can vary within ranges conditioned by the process performance (itself depending on the specific process conditions) and the type of assay used to determine it (affecting Cr only). These ranges were retrieved from our review Andrade et al. [50]. Here, we use the first and third quartiles of Andrade et al. [50] to set "low" and "high" levels for Cc and Cr for all coproducts (Table SI1.2). Combinations of low, mean, and high Cc and Cr were tested for a total of eight new sets of Cc and Cr combinations per scenario.

Since the long-term recalcitrance behavior of biochar is poorly understood due to a lack of long-term experimental evidence in comparison to reported half-lifetimes ranging from decadal- to millennial-scales [72,73], an additional SA was performed on the procedure used to partition Cr within AMG SOC pools. It was performed for the pyrolysis scenario as a representative case of a highly recalcitrant EOM. An alternative partition of the recalcitrant fraction between C_A and C_S was considered. To this end, we considered a remaining C fraction of 75% after 100 years, thus 25% of the initial C is mineralized during the timeframe [50,72]. From the 25%, a fraction is very labile and readily mineralized in the first year (4%) while the remaining corresponds to the mineralizable recalcitrant fraction which is allocated to the C_A pool to be slowly mineralized. More details are provided in SI1.5. The values for pyrochar covered those found in the literature and suggested by the IPCC [74], which proposes that around 80% of C in biochar remains after 100 years for pyrolysis temperatures of 450-600°C. Table SI1.2 summarizes all the combinations explored for the SA.

An excessive application of biochar may be toxic for soil microbiota, which may reduce plant growth and increase CH_4 and CO_2 emissions [52,75]. To avoid this negative effect, an extra scenario was modeled, exporting all the available harvestable crop residues but limiting the soil application rate of pyro- and gaschar to not exceed a total of 50 Mg C ha^{-1} regularly applied over 100 years, as suggested by Woolf et al.[45] to allow char storage in the soil and ensure positive or neutral effects on plant yields. The analysis of alternative storage options for the portion of char not returned to the soil is out of the scope of this work.

Finally, for occurrences where $\Delta SOC_{bio\text{-}BAU}$ (Equation 1) was negative, the portion of retrieved residues from fields was decreased in steps of 25% from its initial 100% value until 0%. These iterations were performed only for scenarios showing negative $\Delta SOC_{bio\text{-}BAU}$ to identify possible compromises between bioeconomy exports and SOC maintenance.

3. Results

3.1 BAU scenario

The BAU scenario predicted a potential decrease of the topsoil SOC stocks by a mean of 2% (Table 2) in the APCUs over 100 years, which represents a C loss of 18 Mt C at the French national scale. Approximately 63% of the simulated areas predicted SOC stocks decrease over 100 years, with a maximum decrease of 27% in some APCUs. APCUs with SOC stock increases may raise their levels by up to 85%, mainly in the Central and Western regions (Fig.S4).

3.2 Bioeconomy scenarios: 100% export over 100 years

Crop residues are already exported for other services in 10% of the areas, making them unavailable for the bioeconomy. Therefore, these areas did not present any change in the bioeconomy scenarios as compared to the BAU scenario. At the national level, the SOC building-up potential over 100 years of each bioeconomy scenario varies greatly; it ranges from -34.9 (molasses) to 774.2 Mt C (pyrochar) (Table 2), a 22-fold difference, reflecting the importance of the coproducts' Cc and Cr parameters, among others.

The highest additional SOC storage, as compared to the BAU, was observed for pyrochar application (+105.5%), while the highest SOC loss was associated with molasses return (-4.4%) (Table 2). It should be highlighted, however, that these decreases and increases are highly variable across the country (Fig. 3), reflecting the large variety of underlying pedoclimatic conditions and cropping practices. This applies to both the BAU (Fig.S4) and bioeconomy scenarios (Fig. 3).

The pyrolysis and gasification scenarios predicted enhanced SOC levels in all the APCUs after 100 years, with the highest potential for SOC sequestration in the Southwestern and Central regions (Fig. 3. a,c). For pyrolysis, SOC stocks increased by over 100% in 57% of the country (Table S2.1), with a national mean SOC stock increase of 105% (+774 Mt C, Table 2). For the gasification scenario, a mean national increase of 43% (+316 Mt C, Table 2) was expected. In 85% of the modeled areas, the consecutive application of gaschar could potentially increase SOC stocks by approximately 80%, as compared to the BAU (max +178%) (Table S2.1; Table S2.4).

The return of hydrochar is shown to ensure SOC sequestration in 88% of the areas (Table S2.4), with a maximum increase of 4%. At a national scale, this scenario represents an average SOC change of 1.1% and a total additional C storage of 8.9 Mt C (Table 2). Unlike pyrolysis and gasification, this scenario indicates potential C losses (up to –1.8%). Digestate may contribute to building up SOC stocks in the North-Central area of France, with SOC stocks shown to slightly increase (max 0.8%) in 50% of the simulated areas (Table 2). Despite the potential SOC storage in this scenario, a mean loss of 0.1% in SOC stocks is expected at a national scale over the timeframe compared to the BAU scenario. For the molasses, the expected SOC stocks after 100 years are lower than in the BAU scenario by a mean of 4.4% at a national scale, representing a potential SOC loss of 35 Mt C (Table 2). The results indicate relative SOC reductions up to 14%, with the highest losses in the Southwest, Northern, and Northeast regions.

For the scenarios depicting SOC losses (i.e., HTL, AD, and 2GEtOH), exporting rates were re-adjusted (75%, 50%). Decreases in the export rates did not influence the overall percentage of areas affected (Table S2.1), thus no lower exporting values were tested. However, the export rate reduction resulted in a lower national SOC loss compared with the 100% export rate for all the remodeled scenarios (Table S2.2).

Table 2. National 100 y SOC changes from the BAU to the bioeconomy ($\Delta SOC_{bio\text{-}BAU}$), in total Mt C and %, at an exporting rate of 100%, at year 2120. Values in % are provided as national averages of all APCUs.

Bioeconomy scenarios	**Total national $\Delta SOC_{bio\text{-}BAU}$[b]**	**Min[d]**	**Max[d]**	**Average national $\Delta SOC_{bio\text{-}BAU}$[e]**	**σ[c]**	**Min[d]**	**Max[d]**
	(Mt C)			**(%)**			
BAU[a]	-17.8	-0.1	0.1	-2.2	14.8	-27.0	84.9
Pyrolysis	774.2	0.0[f]	0.5	105.5	69.3	0.1	409.2
Gasification	315.6	0.0[f]	0.2	43.3	29.3	0.1	177.2
HTL	8.9	0.0[f]	0.0[f]	1.1	0.8	-1.8	4.1
AD	-0.8	-0.0[f]	0.0[f]	-0.1	0.4	-3.5	0.7
2G Ethanol	-34.9	-0.0[f]	0.0[f]	-4.4	2.9	-14.2	-0.0[f]

[a]BAU scenario corresponds to $\Delta SOC_{0\text{-}100}$; [b]Sum of all the modeled APCUs; [c]Standard deviation; [d]Minimum and maximum $\Delta SOC_{bio\text{-}BAU}$ reported over all APCU; [e]Average SOC change for all the modeled APCUs; [f]Value is not zero. More decimals included in Table SI2.2

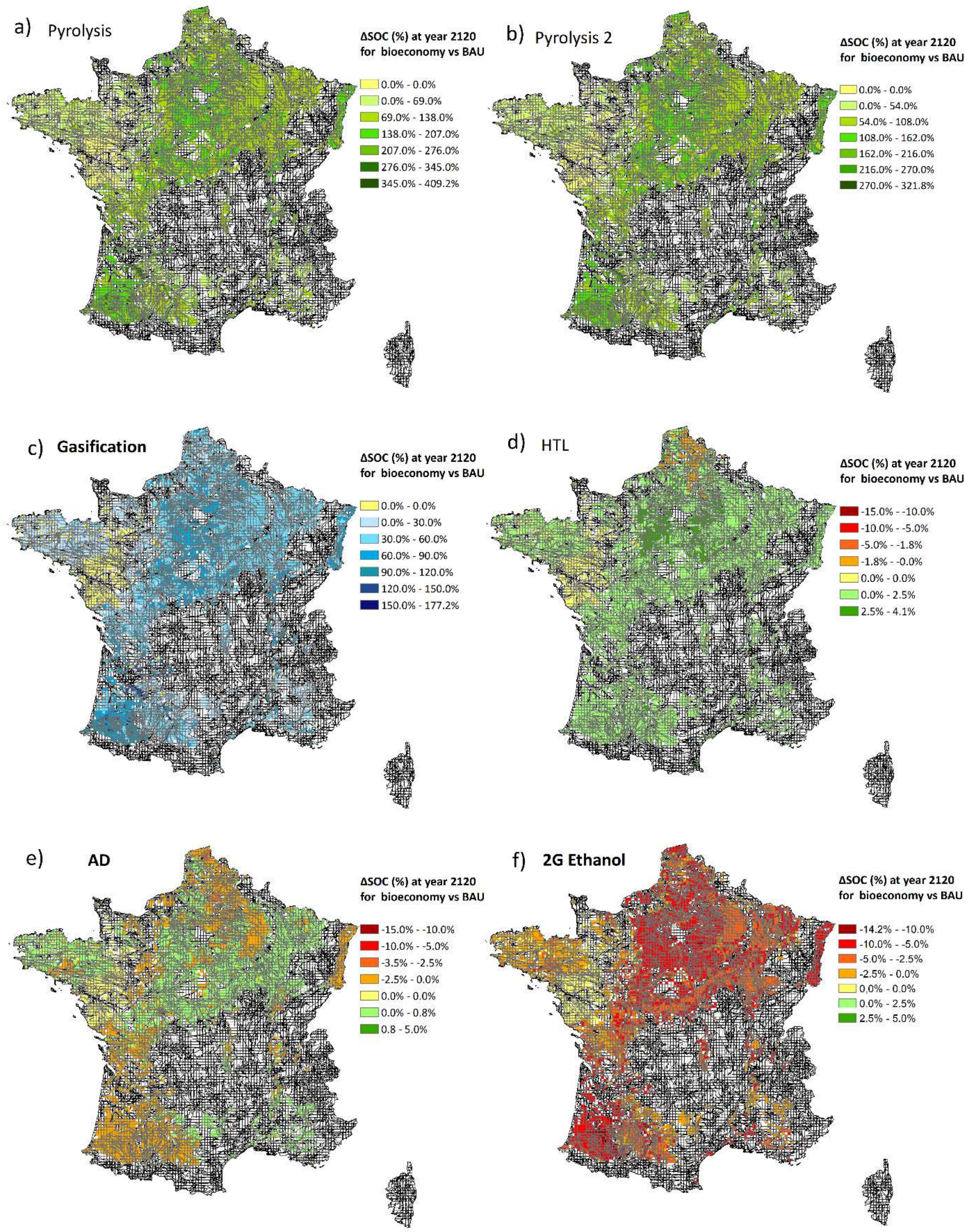

Fig 3. Spatially explicit soil organic carbon (SOC) stocks relative to the BAU scenario (year 2120) if the available harvestable crop residues are used for bioeconomy ($\Delta SOC_{bio\text{-}base}$ %) a) Pyrolysis (with C_S pool of AMG only; default), b) pyrolysis (with C_A and C_S pool of AMG;

sensitivity), c) gasification, d) HTL, e) anaerobic digestion, f) lignocellulosic ethanol. White grids were not included in the simulations.

3.1 Sensitivity Analysis

The SA allowed to evaluate the uncertainty of the potential national SOC changes at the year 2120 due to variability of the Cc and Cr coefficients relative to the mean coefficient value (Fig. 4).

For the pyrolysis scenario, the different combinations of Cc and Cr coefficients affected the additional SOC stocks, ranging between -29% and +30%, equivalent to 549 Mt C and 1009 Mt C (Table S2.3), at a national level. A one-at-a-time test showed the SOC results to be more sensitive to Cc, ranging between -25% and +25%, while Cr ranged between -6% and +5%.

The additional SOC stocks for all the SA tested in the gasification scenario varied by -40% to +36% from that obtained using the mean coefficients, equivalent to 187-431 Mt C. Cr variability contributed to SOC changes between –6% and +5%, while Cc alone affected the results from -36% to +30%.

From these results, it is observed that Cc has the greatest influence on the pyrolysis and gasification scenario, one reason being the greater range of values compared to Cr (Fig.S5, Fig.S7).

For the low recalcitrance scenarios, the uncertainty of the coefficients caused results to vary from C losses to potential additional C storage. The HTL scenario result is affected by -4.8% to +7.6%. High Cc values for any given Cr predict C sequestration in areas that would potentially lose SOC stocks with the mean coefficients. The opposite was observed for low Cc values, which resulted in SOC losses for all the APCUs (Fig.S8). Due to the diverse possible conditions of the HTL technology, the Cc coefficients in this scenario were tested for a broader range (0.12-0.45), which produced a higher effect for Cc than for Cr.

The national SOC change ranges from -16 Mt C to +24 Mt C for the different coefficients in the AD scenario, representing changes of -2 to 3%. The combination of high Cc and Cr resulted in SOC losses in only 0.2% of the simulated areas, compared to 40% for the mean values of the parameters (Table S2.1).

Similarly, lower Cc and Cr values result in SOC stocks decreasing in all the areas (Fig.S9). For the molasses scenario, the combination of maximum and minimum values of Cc and Cr represented a SOC stocks variation of -61% to 48% (Table S2.3, Fig.S10) from the values obtained for the mean parameters, with losses observed in all the APCUs.

The Cr partitioning between the C_A and the C_S pools of AMG (Pyrolysis 2) resulted in cumulated additional SOC stocks of 617 Mt C compared to the BAU scenario by the year 2120 (Table S2.2). This represents a difference of -21% in comparison to a 100% C allocation of the recalcitrant pyrochar to the C_S pool only, with variabilities in the SOC stock results ranging from -39% to -10% for all the SA coefficient combinations (Table S2.3). Albeit the net additional C stored differed for the two Cr allocation methods, the trend observed was the same, with expected SOC increases in all the APCUs. For the Pyrolysis 2 scenario, 36% of the areas predicted SOC increases above 100% (Fig.S6).

If all the harvestable crop residues are exported for pyrolysis or gasification, but only 50 t C ha^{-1} are regularly recycled to the soils throughout the 100 years to avoid the toxic effects of excessive char application, no SOC decreases are observed as a result, on all the APCU.

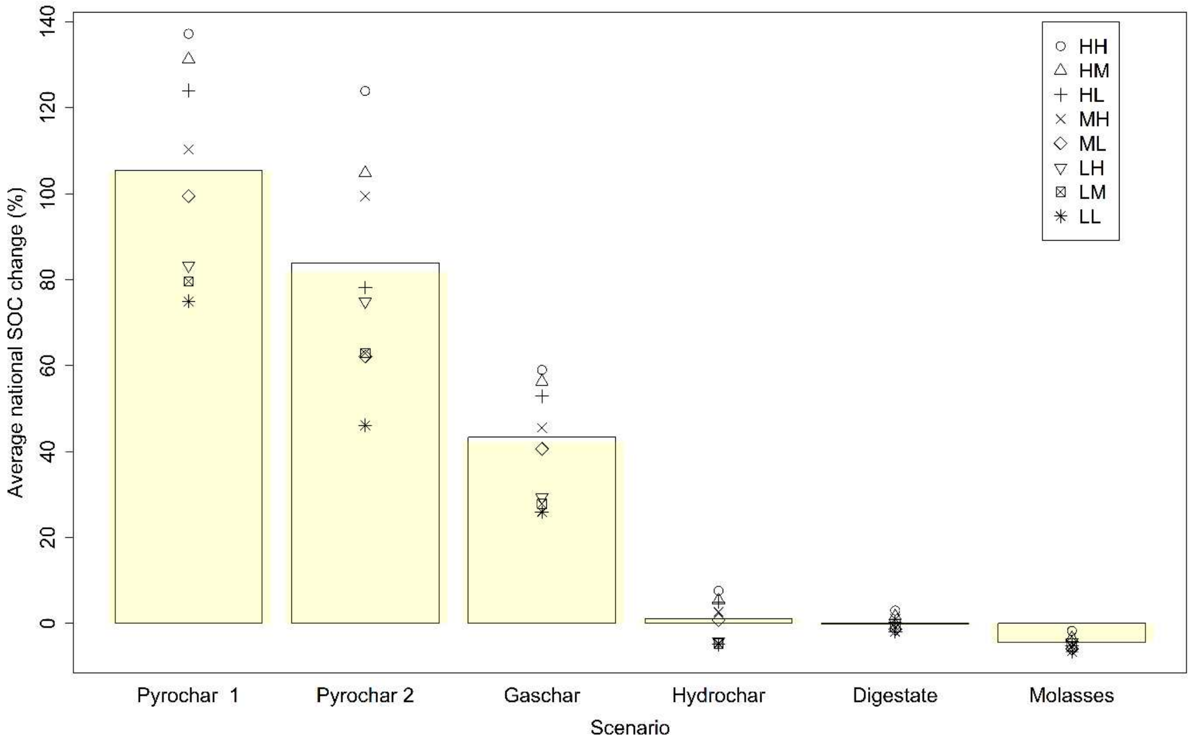

Fig 4. Sensitivity analysis describing a combination of low (L), mean (M), and high (H) Cc and Cr values for each bioeconomy scenario, with an extra scenario for pyrolysis (Pyrochar2) considering an alternative method to partition the recalcitrant fractions into SOC pools in AMG. The bars show the MM (Cc+Cr) value while yellow shades represent the average of all 9 points (SOC at year 2120; in comparison to the BAU) for the different Cc and Cr combinations in a given scenario.

4. Discussion

4.1 Long term spatially-explicit co-products potential for SOC stocks

The BAU scenario, reflecting current practices, predicted a slight average SOC decrease (2% for 100 years) in the simulated areas, which is consistent with the potential prolongation of average decreases in SOC stocks observed over the past decades in temperate croplands in France, Belgium, and Germany [63,76–78]. The simulated decrease is, however, lower than that of 14% obtained by Riggers et al. [79] in German croplands with a multi-model ensemble for the same climate projection (RCP 4.5) and unchanged (current) C inputs for the 2014-2099 period. The BAU scenario predicted SOC losses in around 63% of all simulated areas (Fig.S4). This is in line with the trends observed in Launay et al. [34], where SOC decreases on 55% of the simulated areas (using the STICS model) were observed after 30 years. The regional differences observed can be explained by the influence of the initial SOC stocks, climate, soil, and cropping system characteristics.

In a C-neutral harvesting context, 100% of the harvestable crop residues can be exported for bio-oil or syngas production by pyrolysis or gasification, respectively. Pyrolysis results do compare to those of previous studies. For instance, Lefebvre et al. [19] reported a 127% SOC increase in 20 years in sugarcane fields by replacing sugarcane bagasse and trash with the biochar produced. Woolf and Lehmann [20] found that the export of 50% of maize residues for biochar production, with the subsequent addition of biochar to soils, can increase the SOC stocks by 30-60% over 100 y.

The AD scenario projected a negligible SOC stock increase (up to 0.7%) in 50% of the modeled areas and small SOC losses (up to 4%) in the remaining 40% (the remaining 10% being areas where crop residues are already exported for other uses). Evidence suggests that anaerobic digestion of plant residue little affects SOC stocks on the long term compared to fresh plant-derived C [81]. This was also reported in Thomsen et al. [82]. Bodilis et al. [83] observed a slight decrease in the SOC stocks after digestate application in French croplands, as compared to undigested biomass using AMG. On the contrary, Mondini et al. [28] reported a 2-fold SOC increment after digestate application on Italian lands, compared to undigested crop residues using a modified version of RothC.

The difference between the raw and digested residual biomass lies in the labile C fraction. The removal of the labile fraction reduces CO_2 emissions from digestate compared to the raw feedstock. Besides C, bioavailable nutrients are concentrated in digestate, often in a form that is more assimilable for plants, which provides fertilizing properties [84]. Using digestate as fertilizer can offset the emissions incurred by mineral fertilizer production and application, though excessive application could increase N emissions [85,86]. Areas depicting SOC decreases should therefore be analyzed in detail to determine whether other benefits (energy and nutrient recovery) are worth taking the risk of losing soil C.

The SOC stocks decreased in all the APCUs with the 2GEtOH scenario, which reflects the changed lignin condensation of the biomass exerted by the chemical and enzymatic treatments, allowing the soil microorganisms to decompose the coproducts at a faster rate [13,15,87]. It is associated with increased microbial activity, which may improve fertility and plant growth [88]. Nevertheless, soil application of molasses has been associated with negative impacts on the soil characteristics (e.g., increased salinity and electrical conductivity) and increased GHG emissions in comparison to untreated biomass [13,15]. Our results suggest not exchanging the crop residues provision to soils with bioethanol coproducts if the objective is to prevent SOC losses. More research is required to understand the recalcitrance properties and C content of bioethanol coproducts to harness its potential as a soil amendment.

4.2 Crop residues potential for bioeconomy

In France, it is suggested to limit the harvest of cereal straw to leave a share of 41-96% of the technically harvestable residues on the soil to preserve its agronomic functions [89]. Similarly, ADEME [86] determined that by 2050, only 21% of crop residues could be mobilized for the specific needs of biogas production due to agronomical soil functions and issues related to competitive use. Our results suggest that these thresholds may be too stringent in a C-neutral harvesting context, even for anaerobic digestion, where a 75% harvest (and return) rate imply SOC losses below 1% in 37.5% of the areas (maximum loss of 2.6%, in 2.5% of the areas).

The results of this study demonstrated that the harvest potential is 100% (of the technically harvestable feedstock not already used), unless the residues are to be used for bioethanol (then 0% removal). If to be conservative, we consider export rates of 0% only in the areas where SOC losses are observed with anaerobic digestion and HTL, a reduction of the corresponding crop residue potential of 80% and 3% would be observed, respectively (based on 2021, where the non-exported harvestable crop residues totaled 30.4 Mt DM). Comparing this with the potential of applying a generic 68.5% limit (middle of the above range suggested for France) of residues to be left on land, it involves that between 4 (for anaerobic digestion) and 11 (for pyrolysis and gasification) Mt dry matter of additional crop residues are obtained by applying our framework. This corresponds to an additional supply of 60.4 – 191PJ y^{-1} (for a low heating value of 17.5 GJ t^{-1} DM), the equivalent of the gross electricity generation in Greece and Austria, respectively [90].

Current French cropping systems must increase the C inputs by 42% on average to reach the 4‰ target, while recent works predict a required increase of 283% for Germany [79,91]. However, a decreasing SOC stock trend under a BAU scenario has been identified in this work and others [34,91]. In this context, the management of crop residues, allowing to increase SOC stocks as in the biochar scenarios (pyrochar, gaschar, and hydrochar) and partially in the digestate scenario, could represent alternatives towards the 4‰ goals.

4.3 Strengths and limitations

The scarcity and high variability of data regarding the coproducts C recalcitrance and the challenge of representing long-term effects on real environments based on short-term laboratory studies require caution in the analysis of the results. The main conclusions do not regard the absolute values predicted but the trends related to the sensitivity of the model to the parameters used. We tested a wide range of plausible values for key parameters. The conclusions drawn for each technology can provide insightful decision support with regards to the crop residues' potential for bioeconomy.

The fine granularity of the simulation units assesses the differences among the French croplands, predicting spatially explicit SOC evolutions under each bioeconomy scenario. This approach allows locating areas where coproducts application can build up (or decrease) SOC stocks. Thus, the model can be used to provide advice for resources management for bioeconomy development in specific locations. The framework developed and the modeling approach can be replicated for other regions at different scales, even with less specific granularity, to evaluate the development of crop-based bioeconomy technologies.

Our results partially show the bioeconomy cause-effect link between the usage of the crop residues and their exporting potentials, with different long-term SOC stocks predictions among scenarios. Using coproducts as EOMs soil inputs are expected to modify soil physical, chemical, and biological characteristics in diverse ways. Soil changes can be i) altered net primary production due to changes in the amount and quality of input C and nutrients, ii) addition of extra organic compounds to the soils, and iii) soil microbiota adaptation to utilize the C in the coproducts (this C being structurally different to the one in plant residues) [29,92]. An excessive application of bioeconomy coproducts may alter soil functions which could, in turn, have some environmental impacts. Moreover, the C in the raw biomass is readily available while in the stabilized or recalcitrant matter the C may be unavailable for microorganisms, which could affect soil functioning and fertility. The SA demonstrated that 100% of the crop residues can be exported to increase the bioeconomy provision while at the same time restraining the possible negative effects of biochar, by limiting the application, without affecting the SOC stocks.

Other limitations can be identified in the adapted model and the case studied herein. Changes in soil fertility induced by the coproducts addition were not considered, as well as the potential changes in soil structure and quality due to limitations of the model [52,93]. Besides, nitrogen dynamics (i.e., nitrate leaching and NH_3 emissions) and atmospheric emissions were not evaluated. It was beyond the scope of this work to analyze the overall environmental effects of the different bioeconomy strategies (i.e., accounting for the substituted energy and products by the main bioeconomy products), here focusing on SOC changes only. Similarly, how to prioritize the distribution of each specific crop residue to each bioeconomy technology was not addressed. These considerations, however, need to be assessed in future studies to have a holistic understanding of the environmental impacts of exporting crop residues for each technology.

The future climate trajectory followed the RCP4.5, however, results may vary for different trajectories. The present study considers unchanged cropping systems and crop yields throughout the 100 years. The impact of this hypothesis could be challenged in future works by e.g., using ADEME [86] projections of cropping systems in France, namely one complying with the Factor 4 initiative, and another prolonging the current trends. Factor 4, a national strategy that aims to divide GHG emissions by a factor of 4 by 2050, envisions better agricultural practices and less livestock while the current trends would lead to higher yields for grass, cereal-, and oleaginous crops. These changes in the cropping systems may affect the SOC dynamics and the ability to export crop residues.

We assumed that all cover crops are maintained on soils and all temporary grasslands are exported, while currently ca. 50% of cover crops and 11% of temporary grasslands are being collected at a national scale for anaerobic digestion [86]. This surplus provision of digested feedstock may improve the results obtained for the AD scenario. Moreover, we only considered the changes in recalcitrance for the crop residues digested and not for the co-substrate used. Around 50% of the simulation units involve the presence of manure besides other organic amendments, which could be co-digested, resulting in C inputs decrease but C recalcitrance improvements. Nevertheless, this effect is expected to be of minor importance, in the light of previous works (e.g., Thomsen et al. [82]).

Finally, it should be noted that the SOC losses observed for hydrochar, digestate, and molasses, could be compensated if coupled with other strategies, such as i) redistribution of coproduct from areas showing increased SOC stocks, ii) introduction of specific cover crops, iii) changes in farming management, iv) mix of bioeconomy coproducts return. This was, however, beyond the study scope.

5. Conclusions

This study demonstrated that to maintain long-term SOC stocks, the harvesting potential of crop residues is influenced by the process for which the biomass is destined and is spatially explicit. The partial return of the crop residues C to soils, as stabilized coproducts, was shown to maintain and even increase SOC stocks, in comparison to the levels achieved by just leaving the residues on soils, allowing to provide a greater amount of feedstock to bioeconomy. The study thus confirmed that current practices blindly limiting the potential of crop residues to a stringent threshold unfairly deprive the bioeconomy of an important amount of biomass.

Pyrochar and gaschar were shown, when used as soil C input in exchange for crop residues, to increase SOC stocks in all the French croplands modeled. The HTL scenario predicted SOC stocks increases for 88% of the areas, with a slight loss for the croplands located in the North. The results also indicated that minor SOC gains can be expected through exchanging raw residues for digestate, but only in 50% of the areas, while slight losses were observed on the remaining areas. On the other hand, exporting C from crop residues to be compensated with molasses return was shown to lead to clear losses of SOC stocks in all areas.

By adapting the AMG soil carbon model to consider the recalcitrance of returned bioeconomy coproducts, this study provides an operational tool that can guide, future decisions on the use of crop residues for bioeconomy. However, more research is required regarding recalcitrance, especially of bioethanol coproducts and gaschar, for which studies are scarce and the understanding of the C stability and MRT effects on SOC evolution remain an issue.

Author Contributions

Conceptualization and methodology, C.A., A.A., and L.H.; software and formal analysis, C.A. and H.C.; data curation, investigation, visualization, and writing – original draft, C.A.; writing – review & editing C.A., L.H., A.A., H.C., and E.Z.L; funding acquisition, C.A., L.H., and E.Z.L.; project administration, L.H.; supervision, L.H. and E.Z.L.

Declaration of competing interest

There are no conflicts to declare.

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Data availability

The data that support the findings of this study are openly available in "TBI - Toulouse Biotechnology Institute - T21018" at https://doi.org/10.48531/JBRU.CALMIP/AUEEEJ.

Credits

The graphical abstract has been designed using free icons resources from Flaticon.com (authors: Freepik, Surang, Monkik, Ultimaterm, Kosonicon, Vectorsmarket15, Backwoods, PongsakornRed, Aficons studio, Bqlqn, Smashicons).

Disclosure

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