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The Role of Renewable-Derived Plastics in the Analysis of Waste Management Schemes: A Time-Dependent Carbon Cycle Assessment

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Abstract: Carbon capture and storage (CCS) is an essential greenhouse gas removal (GGR) technology used to achieve negative emissions in bioenergy plants using biomass feedstock (Bio-CCS). In this study, the climate mitigation potential of a novel GGR technology consisting in the production of renewable-derived plastics from municipal solid waste (MSW) refuse has been evaluated. This novel GGR technology allows for carbon storage, for variable periods, in stable materials (plastics), and thus overcomes the technical limitations of CCS. A time-dependent carbon cycle assessment has been conducted based on the Absolute Global surface Temperature change Potential (AGTP) metric. This new method to assess carbon emissions is presented against a traditional life cycle assessment (LCA). The production of renewable-derived plastics proves to be an effective GGR technology for both landfill- and incineration-dominant countries in Europe. The results obtained encourage the implementation of renewable-derived plastics in Integrated Assessment Models (IAMs) to assess their global potential in forecasting scenarios to achieve the ambitious climate change targets set in the European Union. Thanks to this study, a novel approach toward a green and sustainable economy has been established. This study will help to fill the gaps between bioenergy and renewable materials production.

Keywords: plastics; greenhouse house removal; waste-to-energy; dynamic carbon cycle assessment; waste management; Bio-CCS

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1. Introduction

Greenhouse gas removal (GGR) technologies are considered essential for ensuring that a global temperature increase of 2 °C is avoided [1]. Nowadays, the list of GGR technologies is not considered complete, and important research efforts are being made to find more alternative solutions to the existing technologies (e.g., aiming to overcome their technical or economic limitations). Up to now, the GGR technologies identified are (1) bioenergy with carbon capture and storage (BECCS/Bio-CCS); (2) direct air capture of CO_2 from ambient air via engineered chemical reactions (DAC); (3) enhanced weathering of minerals (EW), where natural weathering to remove CO_2 from the atmosphere is accelerated and the products stored in soils, or buried in land or deep ocean; (4) afforestation and reforestation (AR) to fix atmospheric carbon in biomass and soils; (5) manipulation of carbon uptake by the ocean, either biologically (that is, by fertilizing nutrient-limited areas) or chemically (that is, by enhancing alkalinity); (6) altered agricultural practices, such as increased carbon storage in soils (SCS); and (7) converting biomass to recalcitrant biochar for use as a soil amendment [2]. These technologies can be classified according to their level of implementation in Integrated Assessment Models (IAMs) for the modelling of climate change. Therefore, there are common or well-known technologies whose climate impact

is well understood and included in IAMs, i.e., BECCS/Bio-CCS, AR, DAC, and EW; and innovative technologies that require more study or have not been implemented in most IAMs, i.e., biochar, SCS, and manipulation of carbon uptake by the ocean.

Considering the application of GGR in bioenergy systems, BECCS/Bio-CCS is the most relevant technology since it allows for the capturing of the biogenic CO_2 produced in industrial processes using biomass or wastes and helps to avoid its emission to the atmosphere, yielding a net removal of CO_2 from the atmosphere. The IPCC and most IAMs consider this technology to be essential. However, there is a strong social and, in some cases, technical opposition to the implementation of Bio-CCS, making its contribution in IAMs for climate change somehow uncertain [3]. Therefore, additional solutions are needed.

Plastic materials are today part of our lives, and it is hardly imaginable to live without them in the future. Typically, plastics are not considered part of the climate change problem as they only contribute to 4–6% of the global consumption of crude oil [4]. However, efforts have been focused on their production and consumption, with the goal of maintaining the health of our planet. Indeed, the accumulation of mismanaged plastic waste in the environment is estimated to reach around 155–265 Mt/y by 2060 [5]. To mitigate plastic pollution, waste management must be improved to avoid plastic litter. Improving plastic recycling processes could support the shift to a circular economy, reduce the use of fossil sources, provide an incentive to effective waste collection, and contribute to the EU climate neutrality goals. Mechanical recycling has been traditionally extensively employed for the recovery of plastics from waste, but the presence of contaminants in the plastics (coatings, paints, etc.) negatively impacts the mechanical properties of recycled plastics [6]. This drawback has fostered interest in chemical recycling. In particular, the thermal conversion of municipal waste to produce recycled polymers is emerging as a critical perspective technology [7]. The term "renewable-derived plastics" refers to those plastic materials made from renewable sources, such as biomass or wastes. Up to now, renewable-derived plastics have mostly been considered for contributing to a circular economy [8,9], and the European chemical industry has committed to a gradual increase in the utilization of renewable feedstocks, aiming to achieve 25% renewable-derived chemical utilization by 2030 [10].

Renewable-derived plastics also contribute to climate change mitigation because they provide carbon storage in stable chemical structures. Similarly, to biochar, which is used as a soil amendment with the intention to improve soil functions while contributing to climate mitigation due to its long-term storage of biogenic carbon in the soil [11], renewable-derived plastics can offer temporary carbon storage by forming an additional carbon pool [12,13]. Therefore, renewable-derived plastics can be considered as GGR technology.

Geyer et al. commented that humankind has produced around 8300 Mt of virgin plastics [14]. As of 2015, approximately 6300 Mt of plastic waste had been generated globally, around 9% of which had been recycled, 12% was incinerated, and 79% was accumulated in landfills or the natural environment [14]. If current production and waste management trends continue, roughly 12,000 Mt of plastic waste will be in landfills or in the natural environment by 2050. This represents the potential of renewable-derived plastics in the form of GGR technology. However, the real climate and environmental impact of this technology should be analyzed.

The possible environmental benefits deriving from renewable-derived plastics have been analyzed by recent studies, mainly based on the life cycle assessment (LCA) [15,16]. For example, Chen et al. [17] compared 100% bio-based PET bottles versus 100% fossil-based and partially bio-based PET bottles in terms of the LCA. Grabowski et al. [18] performed an extensive review aiming to provide high quality data for the LCA of biopolymers and bio-based materials, identifying an important gap within these data (i.e., the lack of geographical data incorporated in the studies).

The novelty of this study relies on the analysis of renewable-derived plastics as a new GGR technology, including the necessary information for its incorporation into IAMs (at national level). In addition, to our knowledge, none of the studies assessing waste-to-plastic technologies have considered the temporal scale in their analysis. A dynamic assessment of this kind of technology is important because it is necessary to evaluate not only the long-term effects of their adoption on the atmosphere temperature, but also the transient temperature response to know possible undesirable temperature peaks in the short and medium term during the transition to global decarbonatization. This is something that the conventional LCA cannot predict.

With this study we aim to close the gap on the scarcity of information on the potential climate change mitigation of renewable-derived plastics as a GGR technology. In a previous study [19], we evaluated the climate change mitigation potential of waste-derived-fuels as opposed to the current waste management strategies (i.e., landfilling and incineration with energy recovery). In this study, we move forward and compare the climate change impact of the production of renewable-derived plastics from municipal solid waste (MSW) refuse. This option, as opposed to bio-fuel production, stores carbon, for variable periods, in stable materials, and trades the permanency of storage.

2. Methodology

Two systems were defined to assess the climate benefit of producing renewable-derived plastics from MSW: (i) a business as usual (BAU) system, which includes a conventional waste management scheme along with the production of fossil transportation fuels and fossil-based plastics, and (ii) a renewable-plastic (BIO) system, which alternatively produces renewable-derived plastics in an advanced waste-to-energy (WtE) plant producing fuels and plastics using MSW refuse as feedstock. The study was applied to two European countries which exemplify the two main waste management schemes in Europe: (i) Sweden's, which represents the case for Northern and Central Europe, where incineration with energy recovery (heat and electricity) is dominant and landfilling is negligible; and (ii) Spain's, representative of Southern and Eastern European, where landfilling is dominant and energy recovery is focused on electricity production using landfill gas.

Mass and energy balances were performed to obtain the inventory for the carbon assessment of both systems. It was considered that the drop-in renewable-derived plastics are to be disposed of at the end of their lifetime following the same path as fossil plastics in the waste management scheme. Since the different final disposal techniques for waste plastics have an impact on both the climate impact of the baseline case of MSW refuse management and the effective biogenic carbon storage of renewable-derived plastics, cradle-to-grave emissions were considered in this study. The effective greenhouse gas (GHG) removal of the renewable-derived plastic also depends on its time of use, therefore a time-dependent assessment was applied, where the uncertainties associated with the timing of biogenic carbon storage were accounted for. The resulting climate benefit was dynamically assessed and a period of 100 years was considered.

The time-dependent carbon cycle assessment was conducted based on the Absolute Global surface Temperature change Potential (AGTP) metric, and not using a more conventional LCA. The main reason for the authors' decision is that LCA studies consider each environmental impact associated with every stage of the product's life, whereas in this study the authors meant to analyze the climate benefit of the technologies explained above through a carbon analysis. Moreover, the aim of this study is not to study the environmental performance of the biorefinery but to understand where the major benefits in terms of negative emissions are. Therefore, this real case study will serve as an example of the potential of this novel methodology to assess AGTP in GGR technologies.

In Section 2.1 of the methodology the BAU and BIO systems are described, while in Section 2.2 the method applied to assess the GHG emissions of the BAU and BIO systems is explained.

2.1. Definition of the Systems

Figure 1 shows the system boundaries for the BIO and BAU systems analyzed in this study. The size of the systems is independent of the country assessed; only the input composition and the mix of final products are adapted to the country under study. For a fair comparison, both systems were designed to treat the same amount of MSW refuse and produce the same amount and type of products (electricity, fuels, heat, and plastics). For this, in the BIO system it was necessary to import electricity from the grid and heat from the district heating grid while in the BAU system to consider the production of fossil-derived fuels and plastics (Table 1).

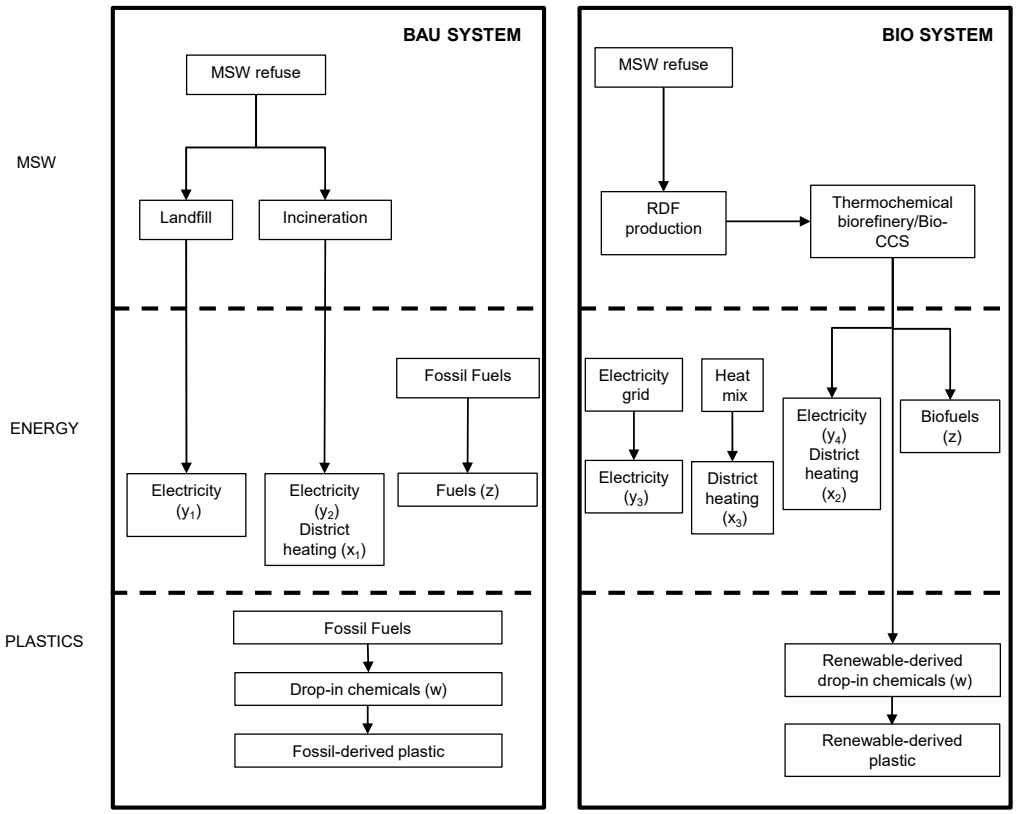

Figure 1. Systems boundaries for the BAU (conventional waste management, and fossil chemicals and transportation fuel production) and the BIO (production of renewable-derived plastics from MSW refuse and balance of electricity and heat, district heating, production from conventional waste management) systems.

Table 1. Shares of renewable-derived plastics, drop-in chemicals, fuels, heat and electricity in the BAU and BIO systems (lower heating value, LHV, basis), as defined in Figure 1.

System	Share (LHV Basis)	Spain	Sweden
BAU/BIO	Drop-in chemicals (w)	24%	13%
	Fuels (z)	23%	12%
BAU	Heat to district heating—Incineration (x_1)	0	54%
	Electricity to grid—Landfill (y_1)	14%	0
	Electricity to grid—Incineration (y_2)	39%	21%
BIO	Heat from district heating (x_3)	0	44%
	Heat to district heating—Biorefinery (x_2)	0	10%
	Electricity from the grid (y_3)	40%	13%
	Electricity to grid—Biorefinery (y_4)	13%	8%

Besides the production of transportation fuels and plastics from fossil fuels, the BAU system includes the conventional MSW refuse management scheme in which MSW refuse is landfilled or incinerated to produce district heating and/or electricity which can be exported to the grid. In the BIO system, MSW refuse is first converted into refuse-derived fuel (RDF), which is transported to an advanced thermochemical biorefinery to be converted into electricity, district heating, and dimethyl ether (DME) and methyl acetate (MA). DME and MA are used to produce drop-in renewable-derived plastics: low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), and poly-vinyl chloride (PVC). These polymers were selected as they account for 60% of the European plastics net consumption [20]. Half of the production of DME is assumed to be used as a substitute for fossil diesel and the other half as a drop-in chemical for LDPE, HDPE, and PP production. All MA is used for PVC production.

The configuration of the thermochemical biorefinery is independent of the region assessed, unlike the impacts associated with the production of heat and electricity in the BAU system (y_1, y_2 and x_1 in Figure 1). Thus, it is necessary to balance the deficit of electricity production from the regional grid and the deficit of heat from the heat mix. In other words, based on Figure 1, $y_1 + y_2 = y_3 + y_4$ and $x_1 = x_2 + x_3$ (Table 1). The functional unit was chosen as 1 MJ of the final product mix (electricity, fuels, heat and plastics), according to the European standard [21], where system expansion is recommended using an energy basis for a system producing fuels and materials.

A description of the modelling of the BIO and BAU systems is provided in Sections 2.1.1 and 2.1.2. As a biomass residue, no upstream emissions (e.g., associated with cultivation, harvesting, and direct and indirect land-use change) are allocated to the MSW refuse for both systems [22].

2.1.1. Modelling of the BAU System

The conventional management of MSW refuse for the selected countries in the study (Spain and Sweden) is shown in Figure 2 [23–25]. In Spain, a higher fraction of MSW refuse is produced, since the recycling ratio is lower than in Sweden. As a result, the RDF composition differs between these countries (Table S1). Heat production is only considered for the Swedish case study since for the Spanish case study heat production is negligible.

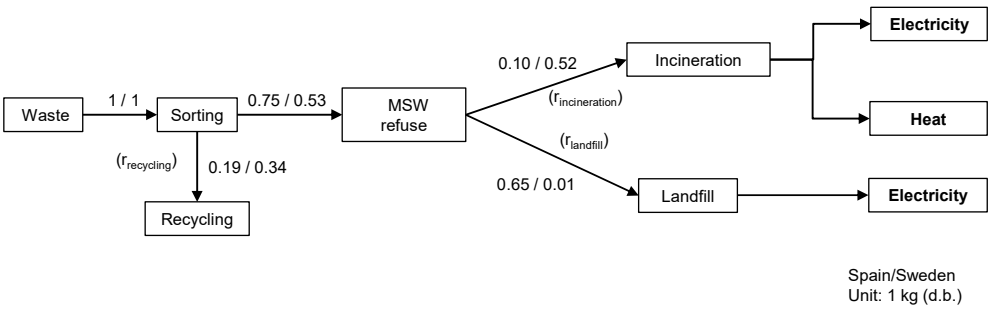

Figure 2. Conventional management of MSW refuse for the selected countries in the study (Spain and Sweden). Final products are shown in bold. Values are in dry mass basis considering 1 kg of MSW refuse as input [23–25].

The GHG impact of the BAU system (E_{BAU}) is calculated by integrating the emissions from the different processes involved (cradle-to-grave) in g of CO_2 eq. MJ^{-1} of the final product mix, (Equation (1)). EBAU is calculated using the emissions associated with landfilling and incineration (waste management) and the impact of the background processes for transportation fossil fuels (EF_{fuel}) and plastics ($EF_{PE/PP}$ and EF_{PVC}) (see emission factors in Table S4, Section 2 of Supplementary Materials). $E_{landfill}$ represents the GHG emissions from the landfill, which depend on the landfilled materials. Although IPCC and EPA establish default values, some parameters are country-specific and even vary between regions [26]. $E_{incineration}$ represents the GHG emissions from MSW refuse incineration

with energy recovery. See Section 2 of the Supplementary Materials (Tables S2 and S3) for input data used in the calculation of $E_{landfill}$ and $E_{incineration}$. In Equation (1), $r_{landfill}$ and $r_{incineration}$ is the fraction of MSW refuse sent to landfill and incineration (Figure 2); and p_{PE} (0.362), p_{PP} (0.204), and p_{PVC} (0.503) is the fraction (on LHV energy basis) of drop-in chemicals converted to PE, PP, and PVC whose production is taken as that calculated in the BIO system to balance plastic production between both systems.

$$E_{BAU} = (E_{landfill} \cdot r_{landfill} + E_{incineration} \cdot r_{incineration}) \cdot (x_1 + y_1 + y_2) + EF_{fuel} \cdot z + (EF_{PE/PP} \cdot p_{PE} + EF_{PE/PP} \cdot p_{PP} + EF_{PVC} \cdot p_{PVC}) \cdot w \quad (1)$$

2.1.2. Modelling of the BIO System

Figure 3 shows the elements of the BIO system and their corresponding carbon flows and pools. The most important element of the BIO system is the thermochemical biorefinery, where firstly the RDF is gasified and converted into syngas. The syngas is cleaned-up by removing impurities harmful for the downstream catalytic synthesis of DME and MA, and then conditioned to fit the composition required in the synthesis by adjusting the syngas H_2/CO ratio and removing CO_2. The captured CO_2 is compressed and sequestered (Bio-CCS) to reduce the CO_2 emissions of the plant. A larger capture of CO_2 would only be possible if the flue gases from the power island were also treated. The biorefinery is designed to maximize the production of DME and MA by means of energy and mass integration, including recirculation of unconverted syngas. A detailed description of the thermochemical biorefinery is provided in Section 3 of the Supplementary Materials.

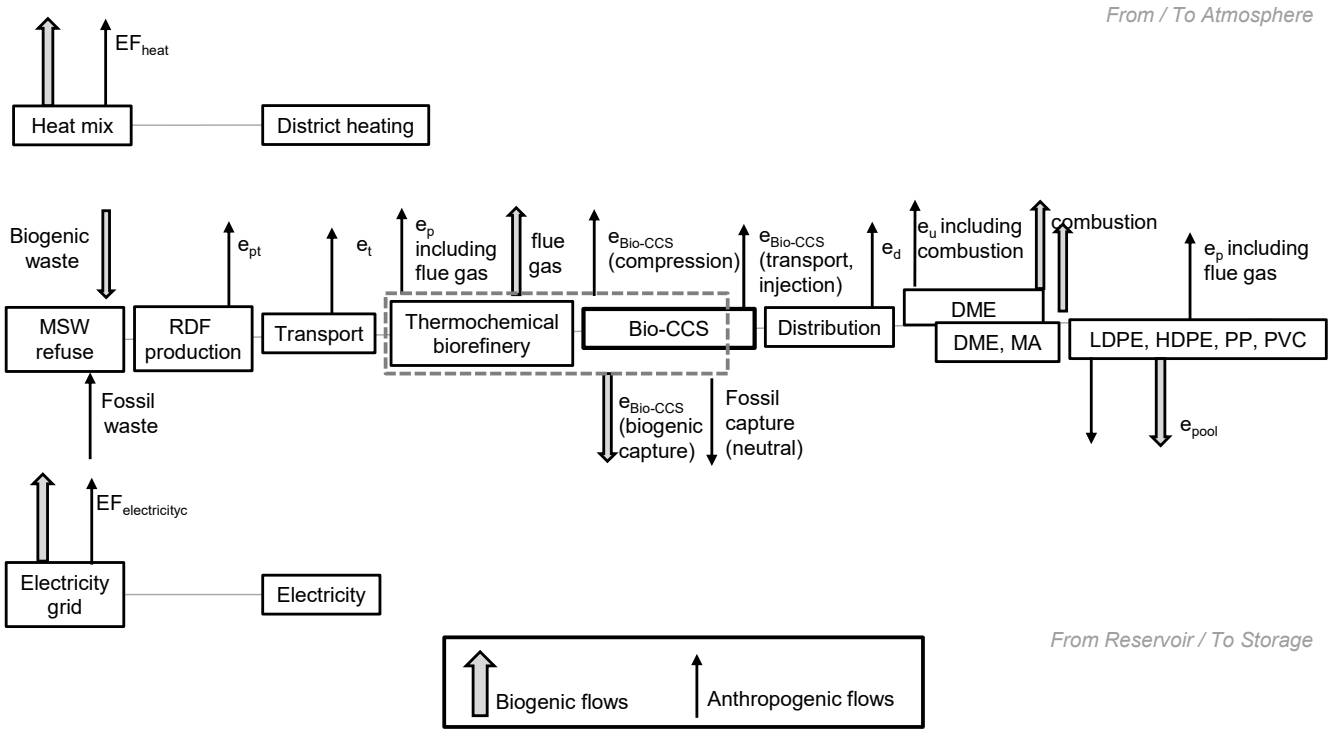

Figure 3. Carbon flows in the BIO system. Biogenic and anthropogenic emissions are differentiated to identify neutral and non-neutral impacts from emissions (upward arrows) and carbon storage (downward arrows). The dashed line envelope reflects the carbon flows from the thermochemical biorefinery.

The carbon flows shown in Figure 3 are emissions to the atmosphere. The biogenic emissions are CO_2 emissions from the biogenic fraction of the MSW refuse (usually more than 50%) and the anthropogenic emissions are non-CO_2 GHG emissions from the biogenic fraction and fossil emissions. Regarding carbon pools, carbon is stored in the form of renewable-derived plastics and as sequestered CO_2 in the thermochemical biorefinery (Bio-

CCS). Since MSW refuse is not fully biogenic, the share of fossil carbon in the renewable-derived plastics was accounted for.

The GHG impact (cradle-to-grave) of the BIO system (E_{BIO}) is calculated by adding up the emissions from the different processes involved (Equation (2)).

$$E_{BIO} = E_{TB} \cdot (y_4 + z + w + x_2) + EF_{electricity} \cdot y_3 + EF_{heat} \cdot x_3 \quad (2)$$

The GHG balance of the biorefinery (ETB) measures the average cradle-to-grave GHG emissions, in g of CO_2 eq. MJ^{-1} of total biorefinery products, associated with the life cycle of the products from the thermochemical biorefinery (Equation (3)). The emission factor for electricity from the grid ($EF_{electricity}$, Table S8) depends on the selected country and, therefore, the average values from the grid mix are used [27]. For heat from district heating, a conservative emission factor (EF_{heat}, Table S8) is taken from the Swedish heat mix based on heat production from waste and biomass combined heat and power plants, as well as from industrial excess heating [28]. There is no heat demand in the Spanish case.

$$E_{TB} = e_{pt} + e_p + e_t + \left(x_{DME} \cdot f \cdot e_{u,fuel} + x_{DME} \cdot (1 - f) \cdot e_{u,chem}\right) + x_{MA} \cdot e_{u,chem} + x_{elect} \cdot e_{u,elect} + x_{heat} \cdot e_{u,heat} + e_{BioCCS} - e_{pool} \quad (3)$$

In Equation (3), f is the fraction of DME used as biofuel (0.5), x_i is the fraction of product i in the biorefinery products (MJ/MJ), e_{pt} is the GHG emissions from pretreatment and RDF production, e_t is the GHG emissions from the transport of the RDF to the thermochemical biorefinery, e_p is the direct GHG emissions from the thermochemical biorefinery, $e_{Bio\text{-}CCS}$ is the net GHG emissions from carbon capture and storage, e_u is the GHG emissions from the use of the final products, and e_{pool} is the biogenic carbon storage in renewable-derived plastics. Details for the calculation of all carbon flows are given in Section 3.2 of the Supplementary Materials. Some considerations on e_{pool} are given in this section.

For the calculation of E_{TB}, the inventory of the thermochemical biorefinery is necessary. Mass and energy balances for the conversion of RDF into electricity, biofuels, and drop-in chemicals (Tables S6 and S7) were obtained by adapting mass and energy balances from a previous study by the authors [29,30] where lignocellulosic biomass was used as raw material instead in the same biorefinery.

The storage of biogenic carbon in renewable-derived plastics (e_{pool}) depends on the type of plastic material and its end-of-life. Whereas landfilled plastic waste is considered in this study a permanent carbon pool, recycled plastic waste is a temporary carbon pool whose permanence depends on the lifetime of the plastic material and the number of recycling cycles achieved. For the considered plastic materials in this study, LDPE and HDPE are assumed to be used for short-time products whose lifetime is under 1 year (e.g., plastic bottles). Thus, 100% of short-life plastics become waste every year. However, PP and PVC present a longer durability and resistance and they are used mostly in the production of long-life products (over 1 year). In Europe, 60% of plastic materials are long-life plastics, whereas 40% are short-life plastics. Every year, 44% of plastics become waste, whereas 56% of them remain in use [31]. Thus, 6.7% of long-life plastics become waste every year. Once plastic materials become waste, the final disposal depends on the region considered. In Spain, 33% of the plastic waste is mechanically recycled, whereas 50% is landfilled and 17% incinerated with energy recovery. In Sweden, 38% is recycled, whereas 59% is incinerated with energy recovery and only 3% landfilled [31,32]. Recycled plastics retain an important fraction of the carbon stored in the original plastic material. Plastic waste going to incineration instead releases the carbon stored as both CO_2 and non-CO_2 GHG to the atmosphere [26]. Since our study has a strategic goal of assessing the climate change mitigation potential of converting waste to plastics as an alternative to current BAU uses, the absorption of CO_2 in the original biomass waste in the MSW refuse cancels out, and we do not attempt to account for it. Instead, the emissions trajectories of biogenic CO_2 are considered in detail for all the systems.

To calculate the equivalent storage of carbon in 1 MJ of plastic produced in one year ($e_{pool,}$ Equation (4)), three variables were defined (Figure 4): the biogenic carbon captured in products still in use and in recycled products, i.e., plastic materials in the market ($e_{material}$); the biogenic carbon captured in plastic waste landfilled ($e_{landfill}$); and the anthropogenic emissions (carbon flow) from incineration (e_{energy}).

$$e_{pool(year\ i)} = \left(e_{material} + e_{landfill} + e_{energy}\right)_{year\ i} \quad (4)$$

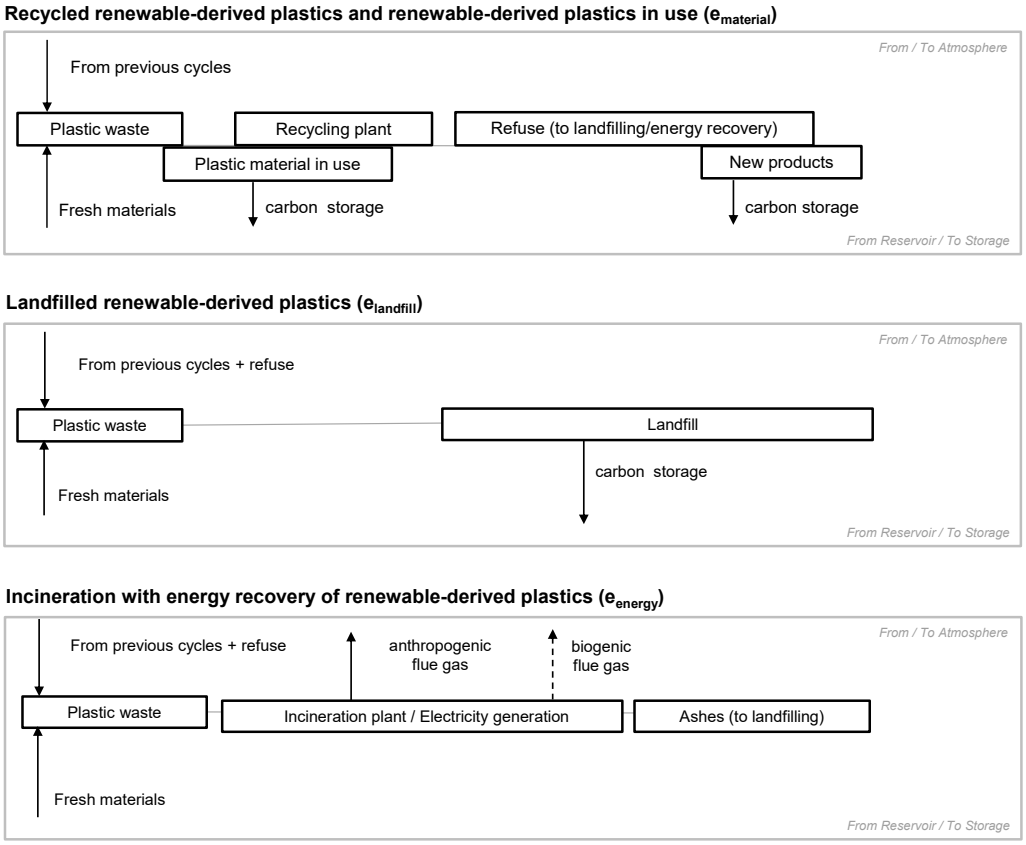

Figure 4. Representation of $e_{material}$, $e_{landfill}$, and e_{energy} defined to calculate the dynamic carbon storage in renewable-derived plastics.

Section 3.3 in Supplementary Materials explains how to calculate the time evolution of $e_{material}$, $e_{landfill}$, and e_{energy} for a pulse of 1 MJ of new renewable-derived plastic and integrate it into the market. This allows us to perform time-dependent assessments of GHG emissions. Figure 5 shows the time evolution of $e_{material}$, $e_{landfill}$, and e_{energy} from a single carbon input in the year 0 by assuming no change in the MSW management system of each country. The carbon input is higher in the case of Spain due to the higher biogenic fraction in the Spanish MSW refuse. The impact of the $e_{material}$ in the carbon pool is higher than the impact of $e_{landfill}$ for the first 15 years in Spain, whereas in the case of Sweden, $e_{material}$ is dominant for 60 years. The high Swedish incineration ratio makes e_{pool} positive from year 20, whereas this is permanently negative in the case of Spain.

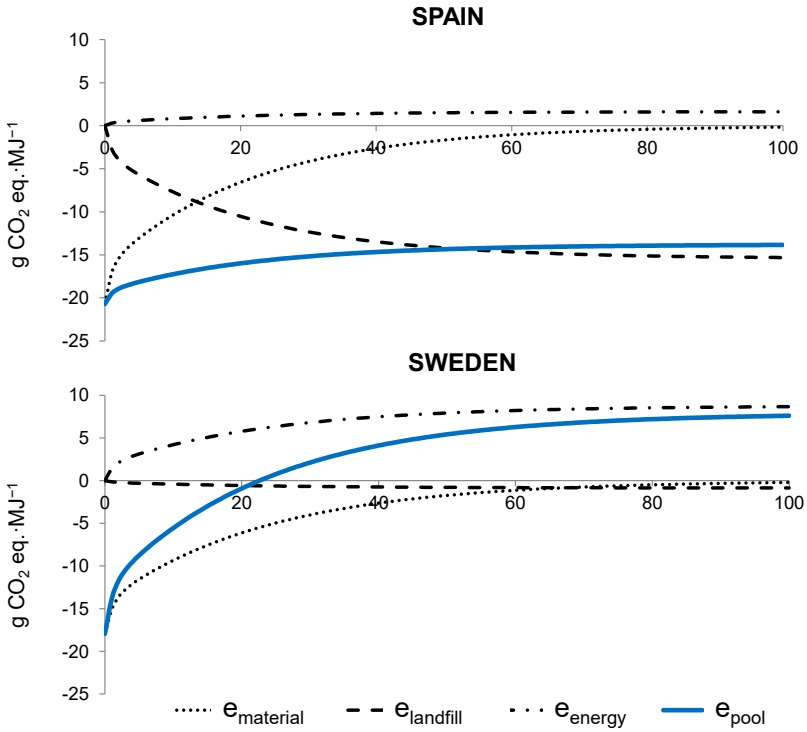

Figure 5. Evolution of the biogenic carbon pool for a single 1 MJ of renewable-derived plastic produced in year 0 (epool) as a function of the carbon flow (e_{energy}) and pools ($e_{landfill}$ and $e_{material}$) for 100 years in g of CO_2 eq. MJ^{-1} of renewable-derived plastics. For the calculations, specific data for Spanish and Swedish current waste management schemes are used (Figure 2).

2.2. Assessment of GHG Emissions

Two scenarios are considered, depending on whether the dynamic nature of GHG emissions and carbon pools are neglected or accounted for.

2.2.1. Static Scenario (Differential GHG Impact)

The differential GHG impact (Equation (5)) assesses the potential reduction in GHG emissions when replacing the BAU system with the BIO system [33], but neglects the dynamic nature of GHG emissions and carbon pools and the evolution of the MSW management system in the selected countries. Thus, for the calculation of the differential GHG impact, the BAU system is considered unchanged for 100 years, and an average biogenic carbon storage in renewable-derived plastics ($\bar{e}_{pool}$) is used for this period (Equation (S10)). Due to these approximations, the differential GHG impact cannot give an accurate comparison of both systems, but it does serve to understand their performance.

$$\text{Differential GHG Impact} = E_{BIO} - E_{BAU} \quad (5)$$

The differential GHG impact is expressed in terms of g CO_2 eq. MJ^{-1} of the final product mix over 100 years.

2.2.2. Time-Dependent Emissions (Differential Climate Impact)

Unlike the differential-static evaluation of GHG impact, in the assessment of dynamic GHG emissions an evolution of the BAU and BIO systems is considered. For the BAU system, the evolution follows legal targets and recommendations set by the EU regulation. The analyzed evolution assumes a progressive banning of landfilling in Europe, a decarbonization of the electricity sector, and an evolution of transportation fuels in Europe. For Spain, the targets set in the national plan are used to define the evolution of the MSW management system for the first 5 years of the period analyzed, i.e., 50% of recycling, 35%

of landfilling, and 15% of energy recovery [34]. From the year 2025 to 2120, the management is assumed to be 1% of landfilling, 65% of recycling, and 34% of energy recovery. As Sweden is closer to the European targets than Spain, the objectives of 1% of landfilling, 65% of recycling, and 34% of energy recovery are assumed in the year 2120. Therefore, the same MSW management scheme for both countries in the long term (including plastic waste management) is assumed. The time-dependent assessment also considers the carbon stored in plastic materials (see Figure 5). In relation to the electricity mix, it is assumed that Sweden would keep constant emissions and Spain would gradually reduce its emissions until both countries reached the same level in 2120. Likewise, GHG emissions from district heating in Sweden would also remain constant. For the BIO system, the dynamic storage of biogenic carbon is incorporated considering the evolution of the waste management scheme, as has been done for the BAU system.

The choice of the climate change metric depends on the aim of the research. The Absolute Global Warming Potential (AGWP) is a cumulative formulation developing the GHG emission method, whereas the Absolute Global surface Temperature change Potential (AGTP) provides an indication of the actual surface temperature change at a given time after GHG emission. The AGTP parameter was used in this study. It is only possible to account for fossil carbon and biogenic non-CO_2 emissions. Therefore, the biogenic carbon stored in renewable-derived plastics (e_{pool}) and from Bio-CCS ($e_{Bio\text{-}CCS}$) was modeled as a negative contribution. Only Well-Mixed GHGs (WMGHGs), including CO_2, CH_4, and N_2O, were considered. The values of the AGTP for the BIO and BAU systems were calculated from the annual emissions of each WMGHG (see Section 5 of Supplementary Materials for details). The Differential Climate Impact (DCI) is based on the AGTP values (Equation (6), giving a direct comparison of the climate benefit in the considered region at a specific time.

$$\text{Differential Climate impact} = AGTP_{BIO} - AGTP_{BAU} \quad (6)$$

For the sake of the reader, a summary of the definition of the systems under study and the assumptions considered in the GHG calculations are gathered in Section 5 (Table S14) of Supplementary Materials.

3. Results and Discussion

3.1. Differential GHG Impact

The differential GHG impact is negative for both Spain and Sweden (Figure 6), so there is a climate benefit from shifting to the conventional MSW management system (BAU) to production of renewable-derived plastics (BIO), considering a static assessment for 100 years. This benefit is larger for Sweden (-194 against -101 g CO_2 eq. MJ^{-1} for Spain) because Sweden has larger emissions associated with the BAU system (E_{BAU}) (183 against 134 g CO_2 eq. MJ^{-1} for Spain), due to the lower landfilling rate and biogenic fraction of MSW than Spain, and lower emissions for the BIO system (E_{BIO}) (-11 against 33 g CO_2 eq. MJ^{-1} for Spain). The emissions of the BIO system (Equation (2)) are those from the thermochemical biorefinery (E_{TB}, Equation (3)) and the consumption of heat and electricity from the grid. The emissions of the thermochemical biorefinery (E_{TB}, Figure 7) are higher for Sweden, again, mainly due to the lower biogenic fraction of the MSW refuse, which results in higher direct emissions (e_t), the main contributor to E_{TB} for both countries. However, Sweden has a very low-carbon electricity grid and heat mix, which compensates for the larger emissions of the biorefinery, resulting in lower emissions for the BIO system. Figure 8 shows a sensitivity analysis of the emissions of the biorefinery and the average biogenic carbon storage.

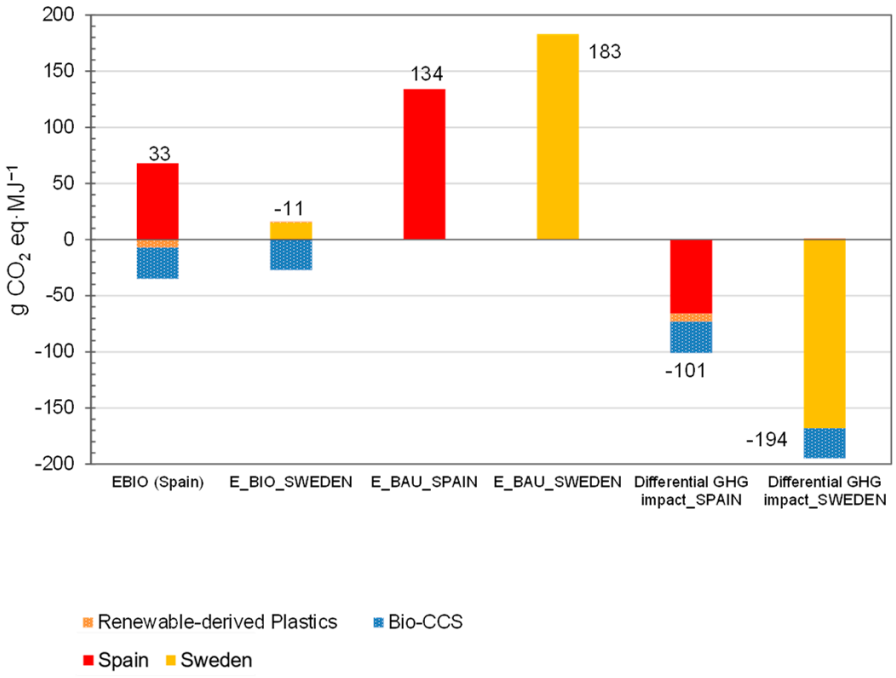

Figure 6. Differential GHG impact based on the emissions from the BIO and BAU systems. The impact of renewable-derived plastics and Bio-CCS is disaggregated. The numbers refer to net values including both renewable-derived plastics and Bio-CCS.

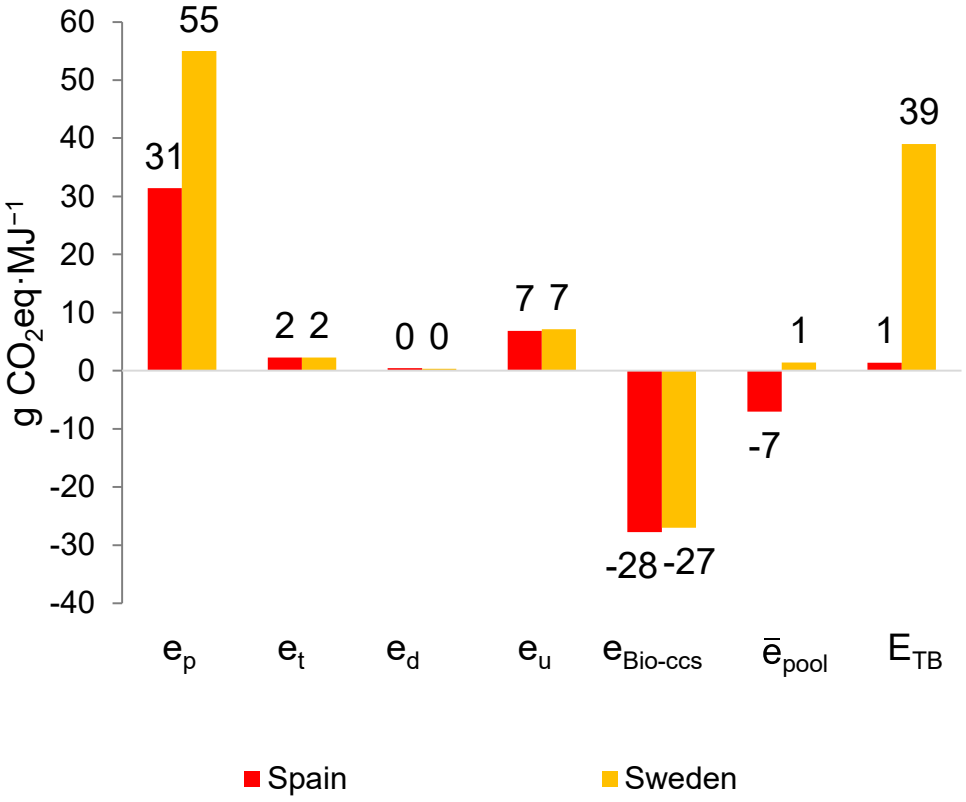

Figure 7. GHG balance of the cradle-to-grave emissions associated with the life cycle of the thermochemical biorefinery using MSW refuse as feedstock. Units: g CO_2 eq. per of 1 MJ of products (sustained production). The variable e_{pt} is not shown since the electricity and heat needed for the RDF production is supplied by the biorefinery. There are no emissions from the distribution of the intermediate products (e_d).

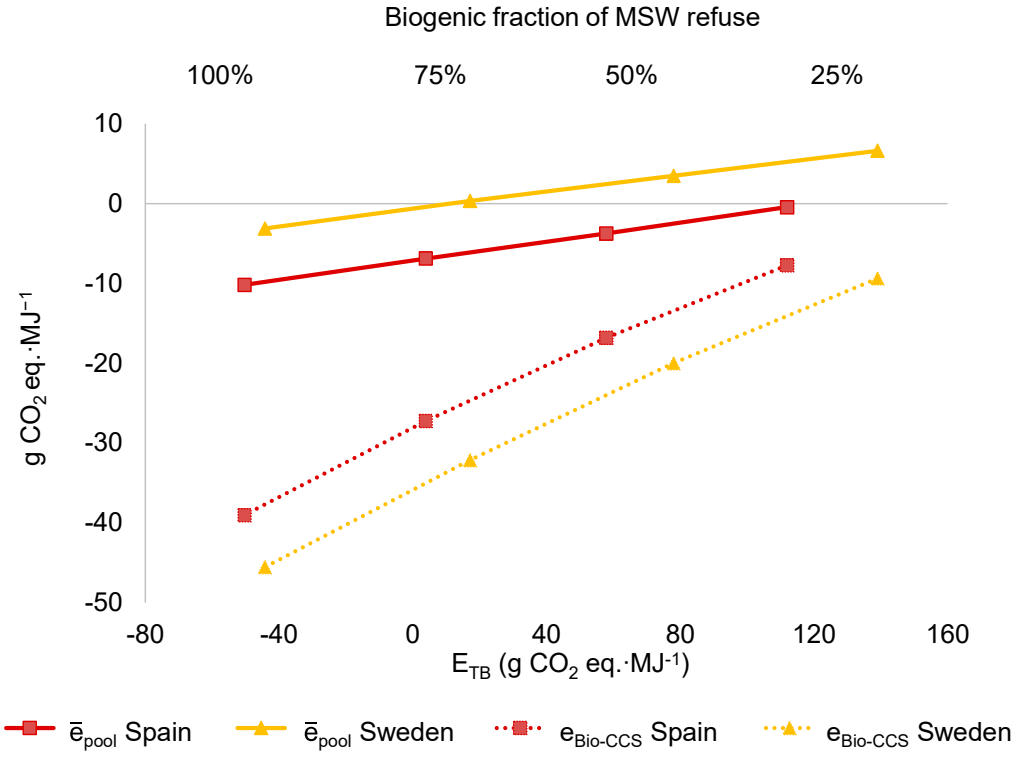

Figure 8. Sensitivity analysis of the emissions of the biorefinery (E_{TB}) and average biogenic carbon storage ($\bar{e}_{pool}$) to the biogenic fraction of the feedstock in Spain and Sweden. The difference in the Bio-CCS process between Spain and Sweden is due to the emissions associated with the electricity consumption during the compression and injection of the carbon. $E_{Bio-CCS}$ in Sweden is better than in Spain, since Sweden has a lower-carbon energy mix.

3.2. Differential Climate Impact (DCI)

The DCI is an appropriate indicator for the analysis of the production of renewable-derived plastics as a GGR technology since it shows dynamic biogenic carbon storage in comparison to the BAU system. Although the results at the end of a time horizon (e.g., 100 years) should be close to that of a conventional assessment of GHG emissions, the DCI allows for tracking the time evolution of the atmosphere temperature. This is particularly interesting when evaluating the climate impact of GGR technologies since unexpected countereffects, such as a temperature maximum, in the short or medium term (e.g., 20–50 years) could appear. Figure 9 shows the DCI (black line) for Spain and Sweden over a period of 100 years. Since the DCI is negative for both countries throughout the whole period, it means that there is a climate benefit as a lower warming of the atmosphere than with the BAU system is achieved. For a net cooling of the atmosphere, the DCI should be below the shaped green area. From Section 3.1, at the end of the period the climate benefit is larger for Sweden on average, but the transient response of the DCI along the period is quite different for both countries. In Spain, there is an inverse response, with a minimum for the DCI in year 17, because of several opposing effects on emissions. As the rate of landfilling decreases with time, emissions from the landfill (especially methane) are avoided [35–37], but the storage of carbon in landfilled long-life plastics decreases. Since methane emissions have a short-term impact on climate change, the initial benefit associated with the reduction in methane emissions disappears after 50 years. In the long term, the increase in the recycling rate tends to increase the storage of carbon due to the reuse of waste plastic, but the most important carbon storage is that of the Bio-CCS incorporated in the biorefinery, which significantly influences the response of the DCI. In Sweden, the transient response of the DCI to a stationary value is smoother since the initial state of the MSW manage system is closer to the European energy targets. As the

incineration share decreases over time, there is a larger storage of biogenic carbon in plastic materials, but this is insignificant to that of the Bio-CCS along the period.

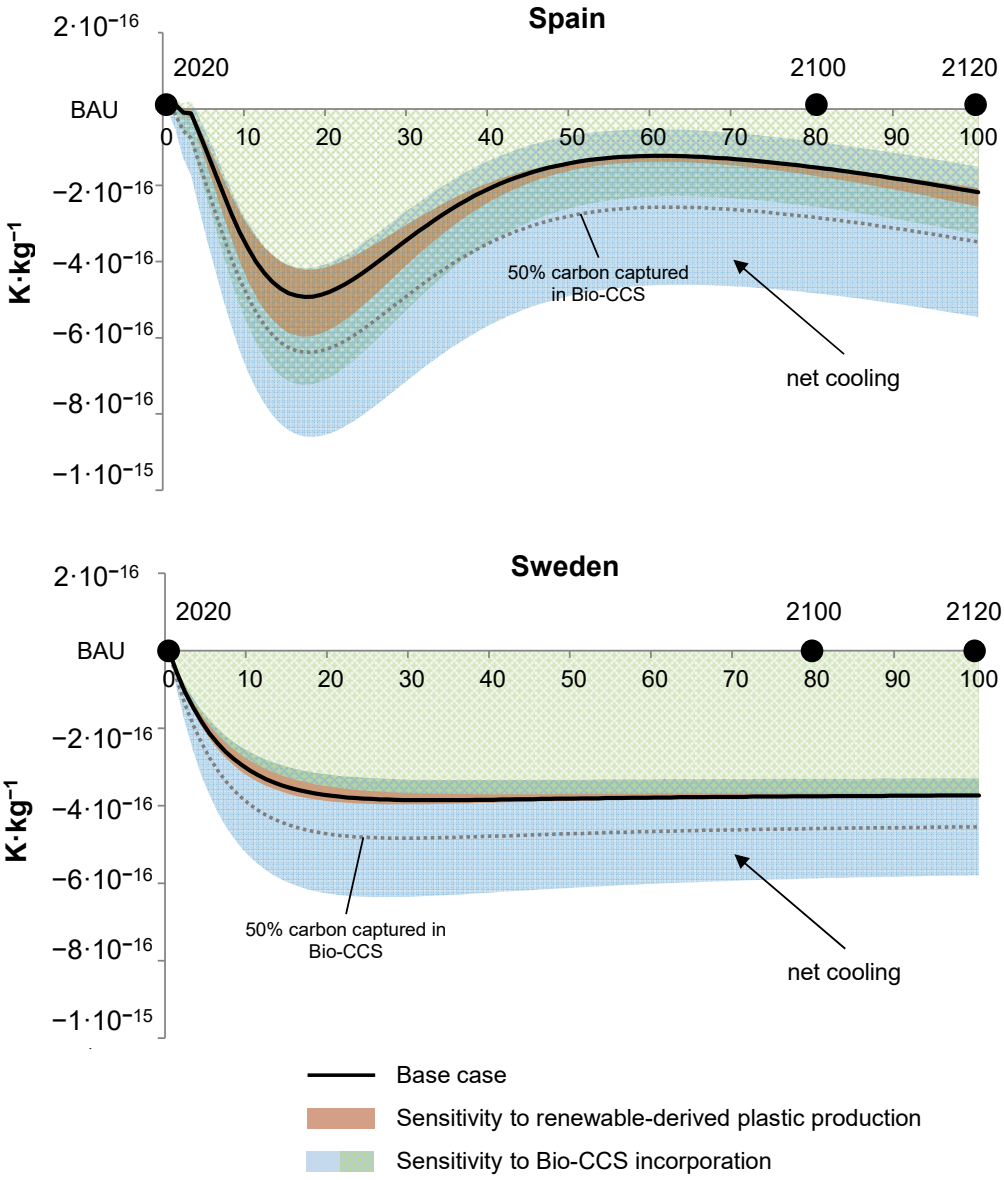

Figure 9. Results of the DCI (variation of atmosphere temperature (K) per kg of MSW refuse treated at year 0) for the base case and sensitivity analysis of the DCI to the share of drop-in chemical used for renewable-derived plastic production and the CO_2 capture rate of the Bio-CCS in the thermochemical biorefinery (Spain and Sweden).

Because the climate benefit depends on the CO_2 capture ratio of the Bio-CCS (carbon captured in Bio-CCS relative to the total carbon emitted by the biorefinery without CCS) and the ratio of DME diverted to the production of plastics in the biorefinery, a sensitivity analysis for these two parameters was performed. The results are also shown in Figure 9. For the base case, the fraction of DME used to produce plastics was set to 50%, and the fraction of carbon captured by the Bio-CCS to 25% (reflecting actual requirements for syngas conditioning for drop-in chemical synthesis [30]). The analysis shows that the DCI is more sensitive to the CO_2 capture ratio than the share of DME for plastic production, as expected from Section 3.1. Only for the first 30 years in Spain is there a comparable impact of the DME share/plastic production and CO_2 capture ratio. A DME share impact-to-plastics ratio of 100% would be equivalent to increasing the capture ratio up to 40% for the

first 25 years. Therefore, the production of renewable-derived plastics has a considerable climate benefit in the medium term in landfill-dominant regions. In the case of Sweden, it is possible to achieve a net cooling of the atmosphere if more than 50% of the DME is used for renewable-derived plastic production. For the case of Spain, it would never be possible to achieve a net cooling, even for the maximum production of plastics. In both countries, a net cooling is possible if the CO_2 capture ratio in the Bio-CCS is larger than 50% in the case of Spain and 25% in the case of Sweden. However, here it is important to distinguish between the feasibility of varying the share of plastic production and the capture efficiency. The share of DME-to-plastics (instead of being used as a transportation fuel) depends on the market and does not involve any technical constraint. However, a higher CO_2 capture ratio (Bio-CCS) would require an increase in both investment and operational costs to capture the carbon present in flue gases and not only the CO_2 available in the syngas (pre-combustion) [38], involving a process more complex [39]. Therefore, it is not likely to have a larger capture efficiency for Spain.

To compare the production of renewable-derived plastics against Bio-CCS as an alternative GGR technology, the DCI of three systems was compared (Figure 10): (i) a reference BIO system (dashed grey line) in which only transportation fuels and electricity are produced in a thermochemical biorefinery from MSW refuse (no GGR technology, dashed grey line); (ii) modification of the reference BIO system by incorporating the production of renewable-derived plastics (BIO system defined in this study without Bio-CCS, only renewable-derived plastics as GGR technology, orange line); (iii) modification of the reference BIO system by incorporating Bio-CCS in the thermochemical biorefinery (only Bio-CCS as GGR technology, blue line). The DCI of the reference BIO system and its modification by incorporating Bio-CCS were calculated in a previous study [19].

As Figure 10 shows, there is a climate benefit after the 100-year period regardless of the region under study and whether a GGR technology is used. For each region, the time evolution of the DCI curves of the BIO systems is similar, which reflects the large impact of the starting waste management scheme on the application of the analyzed GGR technologies. For the first 30 years, the production of renewable-derived plastics and Bio-CCS incorporation have a similar DCI, lower than that of the reference BIO system without any GGR technology. Therefore, both GGR technologies are comparable in the medium term. In the long term, in Spain the increase in the incineration share makes the Bio-CCS a better option (even though the recycling ratio also increases) since it does not rely so heavily on the waste management scheme. On the other hand, in Sweden, since the incineration share decreases over time, there is a larger storage of biogenic carbon in plastic materials, and the production of renewable-derived plastics compares favorably with Bio-CCS incorporation.

3.3. Renewable-Derived Plastics Versus Bio-CCS

For the thermochemical processing of MWS, the climate benefit of carbon capture by producing renewable-derived plastics is like the one gained from not producing plastics but rather by incorporating Bio-CCS in the thermochemical biorefinery, at least in the medium term. Unlike other GGR technologies, the specific climate benefit of renewable-derived plastics and Bio-CCS does not involve a response from the Earth's system to their implementation [40]. However, Bio-CCS presents significant challenges compared with the production of renewable-derived plastics; the latter involving only existing and well-known systems (waste management schemes). Certainly, the largest challenges for CCS deployment are the lack of policy and economic drivers as well as the integration of the component technologies into large-scale demonstration projects (all the individual component technologies are generally well understood and technologically mature if expensive, albeit CO_2 storage still needs further experience at scale, such as with assessing, conditioning, and controlling geological reservoirs for long-term storage and the risk of uncontrolled leakages). Additionally, the lack of understanding and acceptance of

the technology by the public and some stakeholders also contributes to delays in CCS deployment [41].

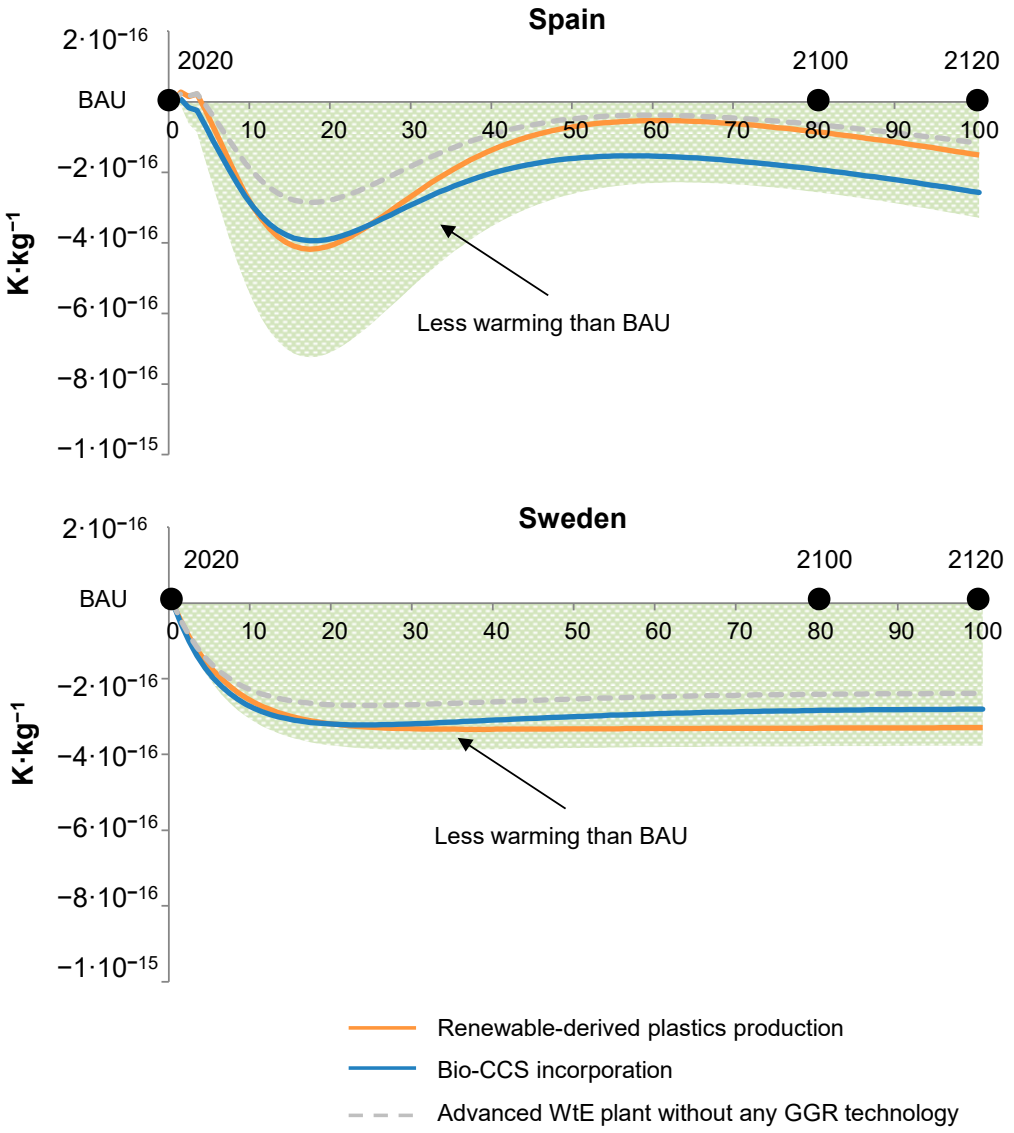

Figure 10. Differential climate impact for (i) a reference BIO system only producing fuels (no GGR technology is incorporated): "Advanced WtE plant without any GGR technology"; (ii) an equivalent BIO system producing renewable-derived plastics: "Renewable-derived plastics production"; and (iii) a modification of the reference BIO system with Bio-CCS incorporation (25% CO_2 capture ratio): "Bio-CCS incorporation".

On the other hand, the limited deployment of renewable-derived plastics into the market might be due to the difficulty in achieving a complete substitution of current fossil-derived plastics. An important challenge is to widen the range of renewable-derived plastic types and possible applications so that they become functionally equivalent to fossil-derived plastics [42]. Other challenges for the deployment of renewable-derived plastics are their right integration into current waste management and the adaptation of manufacturing producers and systems. Only renewable-derived plastic that can be directly introduced into existing and established value chains, infrastructure, and markets have been assessed in the study. Considering regulation issues, the deployment of renewable-derived plastics as a GGR technology may benefit from the EU action plan for the circular economy which foresees economic incentives for packaging producers to put greener products on the

market. One of the main obstacles is the lack of a mechanism to differentiate between fossil-derived and renewable-derived plastics since their composition would be identical. As well as Bio-CCS, the promotion of renewable-derived plastics will provide a platform for the future development of thermochemical biorefineries.

Fridahl [43] performed a mapping of how investments in Bio-CCS are prioritized for the long-term transition to low-carbon electricity generation systems and concluded that Bio-CCS is given a lower priority than renewables but a greater one than conventional CCS. This low preference can be linked to carbon price policy design that incentivizes Bio-CCS and consequently leads to a lack of substantial deployment of this technology. Considering the techno-economic and socio-political uncertainties in the deployment of Bio-CCS, the role of renewable-derived plastics, currently not represented in IAMs, should be assessed in attempts to meet the 2 °C goal. The results shown in this study confirm the climate benefit of renewable-derived plastics as an alternative GGR technology. IAMs could prove the real impact of a substitution of Bio-CCS by renewable-derived plastics in the assessment of future scenarios of climate warming. For the IAMs to include the production of renewable-derived plastics as a GRR, the results presented in this study need to be complemented with their potential at a global scale. In the case of using MSW refuse as feedstock, there would be a large impact in landfill-dominant regions where the potential to avoid methane emissions gives an intense—albeit limited to the first 30 years—climate benefit.

3.4. Climate Change Mitigation Potential of Renewable-Derived Plastics

A simple calculation can be made to estimate the GGR potential of renewable-derived plastics (Table 2). The discrepancies in the values for plastic waste in landfills and world plastic production in 2050 come from the different plastic waste management alternatives, i.e., incineration, recycling, and landfilling. The potential of renewable-derived plastics for mitigating climate change is modest. Compared to the previously evaluated GGR technologies, it is below biochar and increased carbon storage in soils (around 0.5 Gt of biogenic carbon per year, [40]), and significantly below the potential for Bio-CCS (around 3.5 Gt of biogenic carbon per year, [40]).

Table 2. Rough estimation of the climate mitigation potential of a large-scale application of renewable-derived plastics as a form of GGR technology. An average carbon content (mass) of 80% is assumed for plastic materials. The carbon pools for plastics and the fate of plastic residues are taken from [14]. No time-dependent effects have been considered for the calculation of the values in the table. Notes: [a] Each part per million by volume of CO_2 in the atmosphere represents approximately 2.13 Gt of carbon [44].

	Gt of Biogenic Carbon Removed (ppm Reduction), Annually [a]	**Gt of Biogenic Carbon Removed (ppm Reduction), until 2050 [a]**
Europe (all plastic production is renewable-based, only landfill is considered for waste disposal)	0.05 (0.02)	1.5 (0.7)
World (all plastic production is renewable-based, only landfill is considered for waste disposal)	0.27 (0.12)	8.0 (3.8)
All new plastic materials are renewable-based (the amount of plastic waste in landfills from now on until 2050)	-	5.6 (2.6)

4. Conclusions

The production of renewable-derived plastics proves to be an effective technology for greenhouse gas removal (GGR). Its impact as a GGR technology is limited to the total amount of plastics in circulation and the capacity of replacing conventional plastics and, therefore, it would be a minor contributor in the fight against climate impact (similar to biochar). However, the consequences of a large deployment of renewable-derived plastics as a form of GGR technology could change the way waste plastics are managed. For the case of advanced waste-to-energy plants considered in this study, there would be a positive climate benefit compared with current and future waste management schemes in both landfill-dominant and incineration-dominant regions in Europe. The implementation of advanced waste-to-energy plants (e.g., thermochemical biorefineries) producing renewable-derived plastics would have less technical and societal limitations than for Bio-CCS incorporation, would compare favorably in terms of climate benefit in the short and medium term, and would even provide a larger climate benefit in incineration-dominant regions in the long term. The results encourage the implementation of renewable-derived plastics in Integrated Assessment Models to include their global potential in forecasting scenarios to achieve the ambitious climate change targets set in the European Union.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/su15129292/s1, Figure S1: Description of the thermochemical biorefinery as a part of the BIO system. Final products are shown in bold. Values are in dry mass basis considering 1 kg of MSW as input for Spain and Sweden; Figure S2: Carbon balance in the production of renewable-derived plastics from MSW refuse in Spain and Sweden; Table S1: Composition of RDF in Spain and Sweden in % mass basis; Table S2: Input data used in the calculation of the GHG balance in landfilling (BAU system) and results; Table S3: Input data used in the calculation of the GHG balance in the incineration plant (BAU system); Table S4: Relevant data for emissions in the BAU system (biogenic-CO_2 emissions are excluded); Table S5: Demand of selected plastics in Europe, expressed as share of total plastic demand; Table S6: Energy and mass balance of the BIO system for Spain; Table S7: Energy and mass balances of the BIO system for Sweden; Table S8: Relevant emission factors in the BIO systems (biogenic-CO_2 emissions are excluded); Table S9: Emission factors for feedstock transport and final products distribution. Data include empty return trip for a maximum distance of 250 km by truck; Table S10: Parameters used for the calculation of R; Table S11: Parameters used in the calculation of [CO2]; Table S12: Parameters used for the calculation of RF; Table S13: Parameters used for the calculation of AGTP; Table S14: Definition and common-specifics assumptions for both systems and scenarios considered in this study. References [45–63] are cited in the Supplementary Materials.

Author Contributions: Conceptualization, C.A. and P.H.; methodology, C.A. and J.G.; software, C.A.; formal analysis, C.A. and P.H.; investigation, C.A., Á.L.V.P., J.G., J.C. and P.H.; resources, J.G. and P.H.; data curation, C.A. and P.H.; writing—original draft preparation, C.A., J.G., J.C., P.H. and Á.L.V.P.; writing—review and editing, P.H. and Á.L.V.P.; supervision, P.H.; funding acquisition, P.H. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The data presented in this study are available in article and Supplementary Material.

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Conflicts of Interest: The authors declare no conflict of interest.

List of Symbols

e_d	emissions from products distribution (g CO_2 eq. MJ^{-1})
e_{energy}	emissions from energy recovery of plastics (g CO_2 eq. MJ^{-1})
$e_{landfill}$	biogenic carbon storage in landfilled plastics (g CO_2 eq. MJ^{-1})
$e_{material}$	biogenic carbon storage in recycled plastics and plastics still in use (g CO_2 eq. MJ^{-1})
e_{mix}	emission factor from electricity grid mix (g CO_2 eq. MJ^{-1})
e_p	emissions from processing (g CO_2 eq. MJ^{-1})
e_{pool}	biogenic carbon storage in plastics (g CO_2 eq. MJ^{-1})
e_{pt}	emissions from pretreatment (g CO_2 eq. MJ^{-1})
e_t	emissions from feedstock transport (g CO_2 eq. MJ^{-1})
e_u	emissions from the use (g CO_2 eq. MJ^{-1})
r_{energy}	fraction of plastics waste going to energy recovery (%)
$r_{incineration}$	fraction of MSW refuse going to incineration (%)
$r_{landfill}$	fraction of MSW refuse/plastic waste going to landfill (%)
$r_{recycling}$	fraction of plastics waste going to recycling (%)
w_i	MJ of chemicals per MJ of final product mix (i = 1 BAU system, i = 2 BIO system) (%)
x_i	MJ of heat per MJ of final product mix (i = 1 BAU system; i = 2 biorefinery, i = 3 heat mix in BIO system) (%)
y_i	MJ of electricity per MJ of final product mix (i = 1 landfill, i = 2 incineration in BAU system; i = 3 electricity grid, i = 4 biorefinery in BIO system) (%)
z_i	MJ of biofuel production per MJ of final product mix (i = 1 fossil fuels in BAU system; i = 2 biorefinery in BIO system) (%)

Abbreviations

AGTP	Absolute Global surface Temperature change Potential
AGWP	Absolute Global Warming Potential
BAU	Business as usual
BIO	Bioenergy
Bio-CCS	Carbon capture and storage in bioenergy
DCI	Differential Climate Impact
DME	Dimethyl ether
e	Carbon flows
$E_{BIO/BAU}$	GHG impact of the BIO or BAU system
EF	Emission factor (fossil comparator)
EPA	U.S. Environmental Protection Agency
E_{TB}	Global GHG balance of the thermochemical biorefinery
GGR	Greenhouse gases removal
GHG	Greenhouse gases
HDPE	High-density poly-ethylene
IPCC	Intergovernmental Panel on Climate Change
LCA	Life cycle assessment
LDPE	Low-density poly-ethylene
LHV	Lower heating value
MA	Methyl acetate
MSW	Municipal solid waste
PP	Polypropylene
PVC	Poly vinyl chloride
RDF	Refuse derived fuel
SM	Supplementary Material

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