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# Co-recycling of natural and synthetic carbon materials for a sustainable circular economy

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ABSTRACT

Circular economy approaches are commonly depicted by two cycles, where the biological cycle is associated with regeneration in the biosphere and the technical cycle with reuse, refurbishment, and recycling to maintain value and maximize material recovery. This work, instead, presents an alternative vision to the management of carbon-based materials that integrates the two cycles and enables the phasing-out of fossil carbon from the material system. The aim is to investigate the benefits and global potential of a co-recycling system, as an alternative to conventional recycling systems that separate biomass-based materials (e.g., wood, paper) from fossil-based materials (e.g., plastics). Thermochemical recycling technologies enable the conversion of carbon-based waste materials into high-quality synthetic products, promoting circularity and avoiding carbon losses such as carbon emissions and waste accumulation in landfills and nature. Here, the construction and analysis of co-recycling scenarios show how the deployment of thermochemical recycling technologies can decouple the material system from fossil resource extraction. Furthermore, energy use is reduced if pyrolysis and/or gasification are included in the portfolio of recycling technologies. In a decarbonized energy system, deployment of co-recycling can lead to near-zero carbon emissions, while in more carbon-intensive energy systems the choice of thermochemical recycling route is key to limiting carbon emissions.

## 1. Introduction

Carbon-based fuels (mainly fossil fuels but also biomass) account for about 90% of the current global primary energy supply. Carbon is also a building block in a wide range of materials (carbon-based materials; C-materials) used in society. While fossil fuel use is the main cause of anthropogenic greenhouse gas (GHG) emissions, and a transition away from the use of such fuels is essential to limit the global temperature increase to 1.5 °C (IPCC et al., 2018), the production and use of materials such as plastics, cement and steel entail significant GHG emissions (IEA, 2018; Jambeck et al., 2015). The use of biomass-based products can effectively reduce the use of fossil fuels and GHG-intensive materials. There is also scope for substituting existing biobased products with more benign products. For example, cellulose-based textiles can replace cotton which is associated with soil and water depletion, as well as harmful impacts on human health and biodiversity due to excessive use of pesticides and fertilizers (IPCC et al., 2019c; Niinimäki et al., 2020). However, the biomass supply potential is limited by resource constraints (Gerten, 2018) and implications of expanded biomass use for mitigation and other objectives depend on many factors, including soil and climate conditions, biomass type, land management system, scale and pace of deployment, and influence on land use (Calvin et al., 2021). For instance, cropland expansion for energy crop production may cause deforestation, with consequent GHG emissions and negative impacts on biodiversity (IPCC et al., 2019b).

C-materials also entail GHG emissions and environmental impacts at the end-of-life. Landfilling is the most common end-of-life management strategy, and in countries that avoid landfilling the waste is instead often incinerated (Kaza et al., 2018). Much of the material becomes litter, polluting the environment (Kaza et al., 2018). While only a fraction of

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*Abbreviations:* C-material, Carbon material; GHG, Greenhouse Gas; ATCR, Advanced Thermochemical Recycling; CE, Circular economy; CMS, Carbon Material System; MtEn, Material to Energy; MtEco, Material to Ecosphere; CRtM, Carbon Resources to Materials; CCU, Carbon Capture and Utilization; DAC, Direct Air Capture; MTO, Methanol to Olefins; HVC, High Value Chemicals.

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C-material waste is recycled into new products, not all waste can be used today to produce new C-materials. Furthermore, C-materials manufactured from waste often are of lower quality than the original product. For instance, recycled paper pulp is commonly used for newsprint and packaging, which require lower paper grades. Repeated recycling degrades the fibre quality, eventually making the fibres unsuitable for material purposes (Ormondroyd et al., 2016; Van Ewijk, Stegemann and Ekins, 2018), as depicted in Fig. 1. Mechanical recycling of plastics is similarly associated with downgrading (Hahladakis and Iacovidou, 2018), i.e., use in lower-quality products. Thus, today's recycling systems do not achieve full circularity, but merely slow down a linear resource flow that is characterized by losses and quality degradation during reprocessing.

Circular economy (CE) approaches are commonly depicted as two cycles, where the biological cycle focuses on regeneration in the biosphere and the technical cycle focuses on reuse, refurbishment and recycling to maintain value and maximize material recovery (MacArthur, 2019; Mayer et al., 2019). Biogenic carbon flows and resources are part of the biological cycle (Carus and Dammer, 2018; Velenturf et al., 2019). However, C-materials are technical products that can be included in and affect, both the biological and the technical carbon cycles (Kirchherr et al., 2017; Winans et al., 2017). The integration of CE and bio-economy concepts has been discussed in the context of waste management (Teigiserova et al., 2020) and current policy development (Carus and Dammer, 2018; EC, 2018), as well as strategies for reaching targets set in the 2030 Agenda for Sustainable Development and the Paris Agreement on climate change (EFI et al., 2017; 2020). A circular bio-economy emphasizes the use of renewable energy sources and sustainable management of ecosystems, setting limits on biomass usage in society.

Biomass scarcity is an argument for adopting CE principles for the management of biomass that are similar to those for non-renewable resources, i.e., minimize virgin resource use through reuse, recycling and waste avoidance, which also helps to reduce the negative impacts caused by losses, e.g., littering of plastic waste. However, it needs to be considered that reuse and recycling are not always feasible, e.g., when biofuels are used for transport and biobased biodegradable chemicals are used to reduce ecological impacts. Thus, a balanced framework could take its departure in the carbon cycle from a value-preservation perspective and the possible routes for carbon, considering a carbon budget defined by the Paris Agreement and principles for ecosystem protection.

Conventional recycling is commonly investigated and applied separating biomass-based materials (e.g., wood, paper) from fossil-based materials (e.g., plastics, synthetic fibers). In contrast, this work aims to investigate the global potential of a co-recycling system where carbon from the biomass-based and fossil-based waste streams are used as feedstock to produce new synthetic C-materials. Pyrolysis, gasification and combustion with carbon dioxide ($CO_2$) capture and utilization (CCU) are advanced thermochemical recycling processes (ATCR) that allow the recovery and utilization of carbon, regardless of its origin. These thermochemical recycling routes and combinations of them are key enablers for a co-recycling system for converting discarded C-materials, residues, and processing losses into synthetic products of high quality, as depicted in Fig. 1. Thunman et al. (2019) show that by using these three complementary ATCR routes in an existing chemical cluster, it is possible to achieve 100% carbon recovery using today's available technologies. In addition, several authors showed the need and potential of using CCU for a circular carbon economy (Saygin and Gielen, 2021; Meys et al. 2021; Kähler et al., 2021). In all, thermochemical routes can create new opportunities for increasing the recovery of C-materials, promoting circularity, and avoiding carbon losses such as $CO_2$ emissions and waste accumulation in landfills and nature.

In this work, **the production and management of carbon materials (C-materials) are conceptualized and modelled, whereby synthetic and natural C-materials are co-recycled into high-quality synthetic C-materials**, such as plastics and synthetic fibers. The global carbon flows in a co-recycling scheme are described, including ACTR, which provides a novel approach to recycling with a focus on carbon reuse rather than on traditional material recycling. A co-recycling scenario is used to envision how the introduction of these ATCR technologies and changes in carbon flow management facilitate reduced waste generation and decoupling of the Carbon Material System (CMS) from the extraction of fossil resources. Prospective reductions in resource extraction and emissions within the CMS are estimated. In addition, the energy requirement for ATCR is estimated and compared to today's system, including the energy-related emissions given different energy sources.

## 2. Data and methods

In this study, a material flow analysis is set up for the current worldwide CMS that includes global flows of paper, cardboard, wood, plastics and fibers and that accounts for all processes from carbon resource extraction to end of life. A global dataset of C-material products for Year 2018 is used to construct the current CMS, named base case, which is described in terms of carbon mass. A co-recycling scenario is built using the base case, in which the levels of recirculated carbon flows are increased/maximized by exploiting the possibility to convert waste into synthetic C-materials via advanced thermochemical recycling. The parameters defining the base case and the co-recycling scenario can be found in the Supplementary Information (SI), section SI1. All flows are

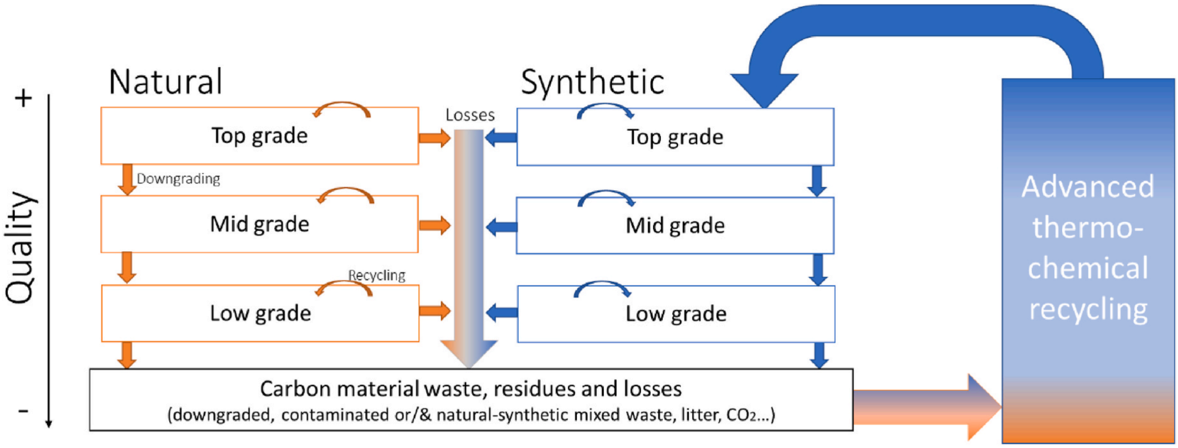

**Fig. 1.** Current downgrading schemes and losses associated with recycling of C-materials (left-hand side) and proposed use of advanced thermochemical recycling to produce high-value C-materials (right-hand side).

reported to the nearest 1 megaton of carbon (MtC).

### 2.1. System definition

Fig. 2 depicts human-induced carbon flows in the CMS and their relationships to the *Ecosphere* and the *Energy System* and Table 1 gives the definitions of carbon flows, resources, products, and losses used in this work. The carbon flows within the CMS are indicated in blue on the right-hand side of the figure. The *Energy System* (left-hand side) includes the energy-related $CO_2$ emissions associated with electricity and heat generation. Since the focus of this work is on the carbon resources used to manufacture material products, Carbon Resources to Materials (CRtM), the *Energy System* description is limited to a generic system that can use Carbon Resources as fuel, as well as other Non-Carbon Energy Sources (e.g., hydro, solar, wind and nuclear).

The CMS comprises three processes: *Extraction & Storage*; *Manufacture*; and *Use & Waste Management (Use & WM)*. *Extraction* refers to the process of mining fossil resources or extracting biomass to be used for material production (CRtM), which is linked to the generation of unused materials (CRtM-Losses). *Extraction* of biomass includes the harvesting and recirculation of part of the carbon back to the forest soil. *Extraction* of fossil fuels includes oil and natural gas, which are converted to high-value chemicals (HVC, i.e., Ethylene, propylene, and aromatics). *Storage* is included in this process, since round wood, chemicals and other materials can remain at a specific location within the CMS and be available for direct use by industry, as well as for future CCU, whereby $CO_2$ could be used for material production (dotted line in Fig. 2). The *Manufacture* process involves all the transformation steps to convert the CRtM into synthetic and natural C-materials. During this process, there is a generation of by-products and waste, which are not used for material production (CRtM-Losses). These flows are either transferred to the *Energy System* for energy generation (MtEn) or simply lost or emitted (MtEco), either way, they end up in the *Ecosphere*. The *Manufacture* process can also assimilate unused C-materials from the *Use & WM* process, such as plastic or paperboard wastes. *Use and WM* consists of the use phase and end-of-life, including the C-materials *Stock* kept in use and post-consumer waste that can be transferred to the *Energy System* for energy recovery (MtEn) or that is lost to final disposal (MtEco). In addition, the waste can be recirculated back to the industry for recycling.

### 2.2. Data

The input data for the existing CMS were collected from the latest available statistics (Year, 2018 in most cases). When data were not available for 2018, earlier statistical data were used, and the values were extrapolated to Year 2018 based on the yearly flow of C-materials produced. The extrapolation was cross-checked with other existing datasets or calculations. The input data and assumptions related to the natural and synthetic C-materials in the existing system are described below and detailed in the SI.

**Table 1**
Definitions of carbon flows, resources, products, and losses.

| | Definition |
|---|---|
| Carbon Resources | Carbon-based resources, including fossil resources (e.g., oil, natural gas) that are extracted from the geosphere and biomass (e.g., wood from forests, agriculture crops) that is obtained from the biosphere. Carbon resources can be used for energy generation or material production. |
| CRtM | Carbon Resource to Material production: these carbon resources are processed into materials (C-materials) or lost during the production process (CRtM-Losses). |
| C-materials | Carbon-based material products manufactured for consumer use. Depending on the manufacturing process, they are classified as synthetic or natural. Synthetic materials (e.g., synthetic polymers and fibers) are manufactured in a synthetic process, using CRtM that can be either biomass or fossil resources. Natural materials retain some properties of the biomass used as the raw material (e.g., wood products, cotton, and paper/paperboard). |
| CRtM-Losses | Losses of carbon from the Carbon Material System as material waste to the *Energy System* (MtEn) and material waste to the ecosphere (MtEco). |
| MtEn | Material to Energy: CRtM used to generate energy in the *Energy System.* |
| MtEco | Material losses to the ecosphere, e.g., waste disposal, combustion without energy recovery, dissipative use and litter or process-related emissions. |

Synthetic and natural material datasets were collected separately. However, C-materials and wastes are often a mixture of natural-synthetic materials, arising from either mixing during collection or blending during the manufacturing process, e.g., a textile with both cotton and polyester or wood with paint or/and varnish.

#### 2.2.1. Natural C-materials

The demand flows for natural C-materials, such as sawn wood, wood panels, and paperboard (including paper, packaging, house and sanitary paper and others), were retrieved from FAO statistics (FAO, 2018), and for natural fibers from the Textile Exchange (TextileExchange, 2019). Conversions between volume and mass units were performed according to the IPCC conversion factors (IPCC et al., 2019a) and natural C-materials were quantified in kg on a dry basis. The conversion to carbon

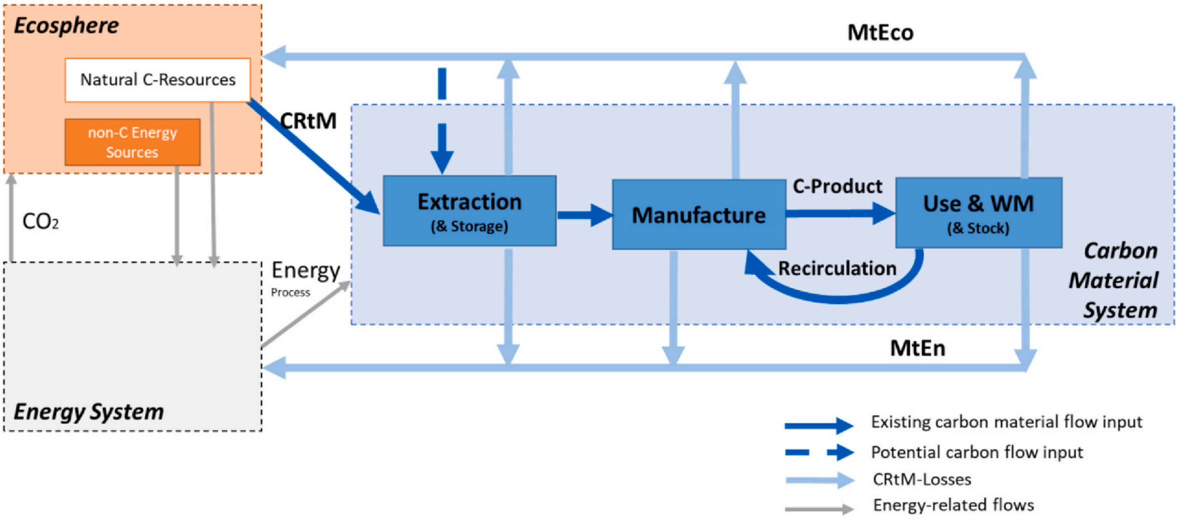

**Fig. 2.** Schematic of the Carbon Material System and associated carbon flows, i.e., the Carbon Resources to Materials system investigated in this work.

flows was calculated according to the carbon content of the products with aggregate value for wood (IPCC et al., 2014; 2019a), and the weighted average for the paper (IPCC et al., 2014; 2019a; Zhou et al., 2014) and natural textiles (TextileExchange, 2019; Zhou et al., 2014) products produced in Year 2018. For more detail see Table S5 in SI.

The amount of biomass harvested for paper and wood products is derived from industrial roundwood (FAO, 2018) material flows with its proportional to forest harvest losses (Born et al., 2014), corresponding to the MtEco in the *Extraction* process (Tables S1–S2). The latter consists of woody biomass that is harvested but not used, so it remains in the forest as logging losses/primary residues (branches, leaves, unsuitable trunks, tree stumps). These residues decompose over time, although part of the biomass is transferred to the soil (Van Ewijk et al., 2018). The CRtM-Losses during the *Manufacture* and *Use and WM* processes were not available for Year 2018, and were extrapolated from earlier material flow analyses for paper (Van Ewijk et al., 2018) and wood (Born et al., 2014). The CRtM-Losses were evaluated by product and differentiated into energy recovery on-site or off-site (MtEn), to stock, recirculated and lost to the environment (MtEco), with the latter including residues burned without energy recovery, landfilled and discarded (Table S4). The carbon flows for paper and wood were cross-checked against European statistical databases (Cazzaniga et al., 2019) that contain more complete yearly datasets. The data for recirculated paper waste were also cross-checked against FAO statistics for Year 2018, i.e., for recovered paper (FAO, 2018), and were calculated according to the carbon contents of the products (Table S6). The recirculated wood flows are those for Europe since wood recycling is scarcely performed anywhere else in the world (Cazzaniga et al., 2019; FAO, 2018). The recirculated natural textile waste has been neglected because its rate of recycling is estimated to be <1% (MacArthur, 2017) (i.e., <0.2 MtC).

### 2.2.2. Synthetic C-materials

The global flows of synthetic C-materials considered here are for polymer resins (PlasticsEurope, 2019) (denoted often as plastics) and synthetic fibers (TextileExchange, 2019). The conversion to carbon flows was calculated according to the carbon contents of the products (Geyer et al., 2017; Zhou et al., 2014). For more detail see Table S5 in SI. The fossil resources extracted have been calculated in a top-down manner on an aggregated level by comparing the input and production levels in the petrochemical industry (IEA, 2018; Levi and Cullen, 2018), together with the CRtM-Losses in the *Extraction* and *Manufacture* processes (see Tables S1–3 in SI). *Use and WM* flows were not available for Year 2018, so they were extrapolated from earlier publications (Geyer et al., 2017; Jambeck et al., 2015; OECD, 2018), evaluated according to product type and differentiated into energy recovery on-site or off-site (MtEn), to stock, recirculated and lost to the environment (MtEco) (UN, 2018). The flows of plastic wastes to incineration, loss, stock, landfilling and recycling were retrieved from the statistical dataset for Year 2017 (Geyer et al., 2017; OECD, 2018), and extrapolated to Year 2018. The synthetic waste that ends up in nature, e.g., in the oceans, is estimated as 4% of the waste generated according to Jambeck et al. (2015). Current CRtM-Losses from collection to recycling through traditional methods were assumed to be approximately 40% (OECD, 2018). The resulting CRtM flows were cross-referenced with previous publications (Kähler et al., 2021; Levi and Cullen, 2018).

## 2.3. Co-recycling scenario

In addition to the base case defining the existing CMS, the co-recycling scenario is analyzed in which advanced thermochemical recycling is implemented in combination with the traditional recycling that is already in place. The co-recycling scenario aims to decouple the CMS from fossil fuel extraction by employing post-consumer waste as feedstock for an advanced thermochemical recycling process that is designed to produce synthetic materials (see Table S7 in SI).

In the co-recycling scenario, the recirculated post-consumer waste is divided into the fractions that can be directly recycled to materials via traditional recycling methods and the fraction that should be recycled via Advanced Thermochemical Recycling technologies. The material recycled fraction, via mechanical and solvolysis methods, has been kept constant (same as the base case) in the scenario construction, while the rest is sent to ATCR. For the fraction of waste going through ATCR, 100% carbon recovery is assumed based on a combination of ATCR technologies (i.e. pyrolysis, gasification and combustion with CCU).

The synthetic C-materials that are lost in the use phase (dissipative use) are assumed to be unfeasible to recirculate, e.g., microplastics produced due to tire abrasion and weathering of marine coating (UN, 2018). Likewise, some paper products may end up in other waste streams (Rogers, 2018; Van Ewijk et al., 2018), and are therefore also assumed to be lost (CRtM-Losses). For instance, sanitary paper is likely to end up in wastewater treatment leaving the CMS (but could be recovered as biogas with digesters). Other synthetic C-materials may be littered or mismanaged in solid waste management systems. These leakage-prone fractions are assumed to be recovered in the co-recycling scenario and used for producing new C-materials.

## 2.4. Resource efficiency

Given the estimated primary resources used and losses (see CRtM and CRtM-Losses in Fig. 2), the resource efficiency of the Carbon Material System has been evaluated for the base case and the co-recycling scenario. Two ratios are calculated: the resource utilization ratio, representing how many tonnes of C-materials are produced per 1 tonne of primary resources (expressed as $t_{\text{C-material}}/t_{\text{CRtM}}$); and the resource loss ratio, describing how many tonnes of resources are lost in the CMS per 1 tonne of C-products (expressed as $t_{\text{CRtM-Losses}}/t_{\text{C-material}}$).

## 2.5. Energy demand

The energy demand and carbon emissions are evaluated from a theoretical standpoint and detailed only for synthetic C-materials, since the natural C-materials side is assumed to remain the same in the base case and the co-recycling scenario.

The energy needs for producing synthetic C-materials consists of the energy in the material feedstock ($\text{Energy}_{\text{CRtM}}$) and the energy demand of the process ($\text{Energy}_{\text{process}}$). This energy comes from virgin resources (CRtM) and the energy system (Energy), respectively, for the base case (see Fig. 2). In the co-recycling scenario, the energy in waste is also recirculated together with the carbon recirculation. To evaluate and compare the energy efficiency between the base case and the co-recycling scenario, equation (1) is used:

$$\varepsilon = \frac{Energy_{synthetic\ C-materials}}{Energy_{total} + Energy_{synthetic\ C-materials}} \tag{1}$$

where: $Energy_{total} = Energy_{process} + Energy_{CRtM}$

Equation (1) takes into consideration the chemically bound energy in the synthetic C-material ($\text{Energy}_{\text{synthetic C-material}}$), since it corresponds to the energy needed for the C-material to be replaced in the system. For instance, if 1.6 units of total energy are provided to produce 1 energy unit (e.u.) of C-materials, in a linear system this 1 e.u is lost, and to replace it, an additional 1.6 e.u. is needed. In a circular system, the C-material energy is recirculated back to the system to replace itself 1:1 and only the energy for the recycling process is needed.

## 2.6. Technologies for co-recycling

As described before, this work considers three possible technology pathways for the co-recycling scenario: pyrolysis, gasification and combustion with CCU (see Thunman et al. (2019) for a summary of these technologies). These will yield different energy demands of the advanced thermochemical recycling (ACTR) in the co-recycling scenario. To provide a first estimate of the energy demand required by a CMS that incorporates thermochemical recycling of mixed waste, the energy demands for the three technology pathways are estimated. The energy demand of a realistic co-recycling system via ATCR is expected to fall within the boundaries of the least and most energy-intensive technology pathways, as a combination of all three technologies will be needed to treat the mixed wastes while achieving circularity. As a theoretical example, the energy efficiencies for these three technology pathways are provided in Table 2, for the case of polyethene production and based on heat balances reported in Thunman et al. (2019). A visual exemplification is included in Fig. S2 in the Supplementary information.

The three technology pathways are labelled as *Low, Medium and High,* which refer to the least to most energy-intensive routes for thermochemical recycling. The *Low* pathway is defined as pyrolysis of olefinic plastics with separation of the products in a heat integrated process. Pyrolysis consists of the direct recovery of the monomers and valuable molecules from the original material through thermal cracking. In the pyrolysis pathway, the chemical structure of the material is partially preserved, requiring less energy than gasification and combustion with CCU, but the structure of the feedstock polymer becomes important. Therefore, this case is strictly theoretical for a 100% carbon recycling case, as later discussed. The *Middle* pathway refers to the gasification route, where the feedstock is thermally decomposed into syngas (mainly $H_2$ and CO) that is used for the synthesis of methanol, which is then reacted into olefins via methanol to olefins (MTO). The *High* pathway involves complete combustion of the feedstock, with $CO_2$ capture followed by the synthesis of methanol, using pure hydrogen from electrolysis to adjust the composition of the syngas. A similar MTO process to that applied in the gasification route is used in this case and it is assumed that there is no heat integration between the involved processes. The potential energy demand for the three technology pathways is summarized in the Supplementary Information Table S10.

As shown in Table 2, gasification and combustion with CCU are less feedstock-dependent than the pyrolysis pathway, allowing the use of mixed and contaminated plastics and other types of carbon-rich wastes, but at the expense of higher energy demand and more extensive processing. In the results, the energy demand estimations for each pathway are compared to the energy demand of the base case, which is normalized to 100% (see Table S10). This normalized value indicates the potential energy efficiency of the co-recycling system in relation to today's situation. Nevertheless, in reality, the efficiency will depend on the waste available and the extent of heat integration.

### 2.7. Carbon emissions

The energy and carbon resources are highly interlinked and, thus, the potential change in carbon emissions when implementing ATCR is also assessed in this work. The carbon emissions have two components, the C-materials related emissions and the energy-related emissions (as depicted in Fig. 2). Todays' CMS energy-related emissions from fossil fuel extraction to produce synthetic C-materials were taken from existing datasets (Levi and Cullen, 2018; Saygin and Gielen, 2021). The combined material and energy carbon emissions of the base case are normalized to 100% and compared to the potential changes in the carbon emissions of the co-recycling scenario presented (see Table S12).

**Table 2**

Summary technology pathways studied and their characteristics (process, feedstock, energy efficiency and main form of energy demand).

| Technology pathways | Processing to olefins | Possible feedstock and waste streams | Energy efficiency ($\varepsilon$) | Main form of energy |
|---|---|---|---|---|
| Base case | Cracking and separation | Fossil fuel | 38% | Heat |
| Low | Pyrolysis and separation | Homogenous PE,PP & PS | 60% | Heat |
| Middle | Gasification, separation and synthesis | Mixed waste | 45% | Heat |
| High | Carbon capture, $H_2$ production and synthesis | Mixed waste | 28% | Electricity |

The potential carbon emissions in the co-recycling scenario consist of the avoided carbon losses by recirculating waste streams and the energy-related emissions. The potential energy-related emissions depend on the energy demand of the ATCR technology chosen (i.e., pyrolysis, gasification, and combustion with CCU) and the energy mix of the *Energy System.* In the results, the potential carbon emissions for the three technology pathways are presented for six types of energy sources: coal, gas, biomass, hydro, wind, and solar PV. Data on heat and power efficiencies as well as carbon intensities ($CO_{2\text{-eq}}$/kWh), including direct (combustion) emissions and emissions associated with infrastructure and supply chains, are based on Schlömer S. (2014) and tabulated in Table S11. For bioenergy, two cases are included. Both cases include emissions associated with infrastructure and supply chains. In one of the cases, also $CO_2$ emissions from biomass combustion are included to show the total amount of carbon emitted compared to the current situation and to clarify the dependence on carbon as a resource. However, the fundamental difference between fossil and biogenic carbon: the combustion of biomass emits $CO_2$ that was previously sequestered from the atmosphere and will be sequestered again if biomass is sustainably sourced (Cowie et al., 2021). Thus, this combustion emission cannot be used as an indicator of the influence of bioenergy systems on the atmospheric $CO_2$ concentration. Finally, nuclear and CCS routes are also summarized in the SI, for comparison, but not depicted in the results section.

## 3. Results

### 3.1. Base case: current Carbon Material System

An overview of the global CMS for Year 2018 is shown in Fig. 3a, with further details provided in Table S7 in Supplementary Information. The amount of carbon in all the C-materials manufactured globally in Year (2018) was about 745 MtC, with approximately 425 MtC in natural C-materials (220 MtC in wood products, 185 MtC in paper and cardboard products, and 20 MtC in natural textiles), and the remaining 320 MtC in synthetic C-materials (280 MtC in plastics and 40 MtC in synthetic fibers).

About 1090 MtC of virgin CRtM were needed to produce the C-materials, corresponding to a resource utilization ratio of about 0.7. Of the 420 MtC of oil extracted, 85 MtC were lost during the transformation processes. Of the 670 MtC of biomass harvested, 530 MtC were extracted and 140 MtC remained in the forest as logging losses/primary residues.

In addition to the 225 MtC of CRtM-Losses experienced during the *Extraction* process, 230 MtC were lost during the *Manufacture* process, representing 200 MtC of losses during the *Manufacture* of natural C-materials and 35 MtC of losses during the production of synthetic C-materials. Of the natural C-materials losses, 110 MtC were sent to energy production (MtEn), mainly to the pulp and paper industry to meet internal process energy needs and some as wood byproducts used as fuels, and the remaining 90 MtC were lost to the environment (MtEco).

The amount of carbon in post-consumer waste was estimated at 475 MtC. Of this, 235 MtC were post-consumer waste of natural C-materials, of which 45 MtC were incinerated with energy recovery and 95 MtC were recirculated to produce new C-materials. The remaining 95 MtC were lost to the environment (MtEco). Around 80% of the recirculated post-consumer waste consisted of paper and paperboard products, which are commonly used for producing newsprint and packaging materials, for which lower quality feedstocks can be used (Rogers, 2018; Van Ewijk et al., 2018). For synthetic materials, the post-consumer waste was 240MtC and most wastes ended up as MtEco (170 MtC), followed by MtEn (50 MtC), and only 20 MtC were recirculated back to

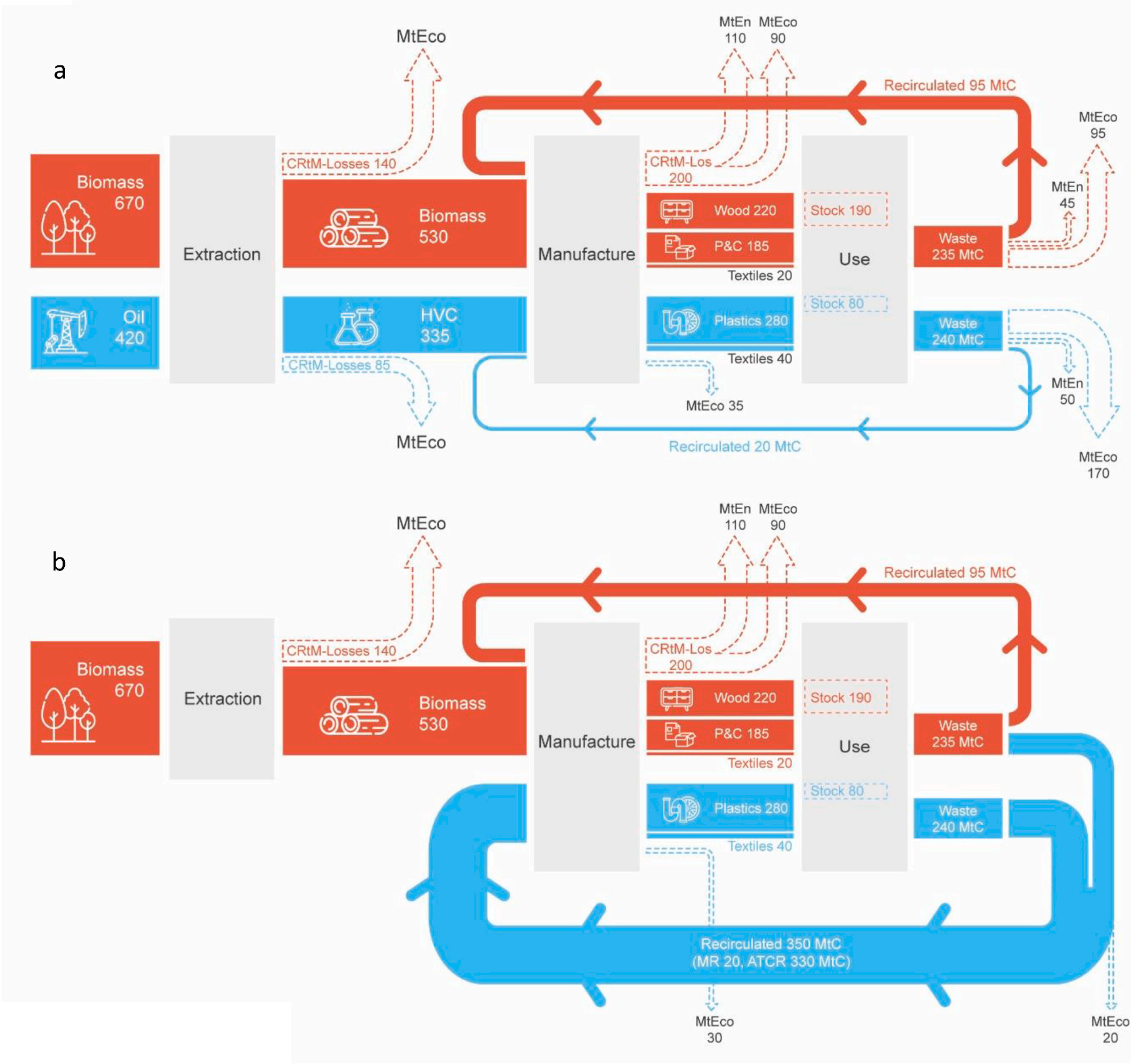

**Fig. 3.** The carbon flows (a) in the Base case (current Carbon Material System) and (b) in the Co-recycling scenario. All units in MtC. P&C: paper and cardboard, MR: mechanical recycling, ATCR: Advanced Thermochemical Recycling. Values are rounded.

the *Manufacture* process, yielding approximately 15 MtC of synthetic products. Almost all the recycled synthetic C-materials ended up in lower quality products, with only 2% found in materials of similar quality to the original material (Geyer et al., 2017; MacArthur, 2019). In total, about 815 MtC were lost from the CMS, which is similar to the total produced C-materials (losses ratio of ≈1). Of the total CRtM-Losses, 205 MtC were combusted for energy generation (MtEn). About 610 MtC ended up in landfills, were lost as process emissions, or simply became litter in the environment (MtEco).

### 3.2. Co-recycling scenario

The co-recycling scenario shows how the CMS can be decoupled from the extraction of fossil resources, thereby avoiding the current extraction of about 420 MtC CRtM for the production of synthetic C-materials, as well as the associated CRtM-Losses of 85 MtC. Thus, the total extraction of CRtM is reduced from 1090 MtC to about 670 MtC. This 670 MtC consist exclusively of biomass, which is the same amount as is currently used. As a result, the resource utilization ratio improves to 1.1.

The results (Fig. 3b) show that the natural C-materials part of the system remains unaffected, and that the post-consumer waste is used to substitute for fossil CRtM as the feedstock for manufacturing synthetic C-materials through advanced thermochemical recycling. It should be noted that, as stated in *Section 2.3*, part of the post-consumer waste has a dissipative nature and some of the streams end up in other waste streams; those streams account for approximately 20 MtC (15 MtC natural and 5 MtC synthetic C-materials) and are assumed to be unrecoverable. Thus, 350 MtC of waste are recirculated to produce synthetic C-Materials, 20 MtC are used in the existing mechanical recycling, and 330 MtC undergo advanced thermochemical recycling (accounting for 215 MtC of synthetic C-material and 115 MtC of natural C-material waste).

Overall, the estimated total CRtM-Losses are 390 MtC (resource losses ratio of ≈0.5), which is approximately half of the CRtM-Losses for the base case. The carbon flows that go directly to energy recovery also differ, with only 110 MtC being sent to MtEn compared to 205 MtC in today's system.

### 3.3. *Energy demand and carbon emissions*

The potential reductions in energy demand of the three technology pathways, compared to that of the base case, are depicted in the left-hand chart in Fig. 4. As shown, the demand depends on the energy required for the chosen ATCR technology pathway. For the *High* pathway, combustion with CCU, the energy consumption of the co-recycling system is estimated to be 161% of the base case energy demand for oil extraction and HVC production. For the *Low* pathway, pyrolysis, the energy demand potential is 44% of the energy demand for producing synthetic C-materials from fossil resources. For the *Middle* pathway, gasification, the system will require 81% of the base case energy demand.

As depicted in the right-hand chart in Fig. 4, the potential change in emissions for the synthetic C-materials manufacture depends both on the energy demand of the ATCR and the energy source. As expected, the highest reduction potential of the carbon emissions requires carbon-free options such as hydro, wind and solar reaching 1–10% of the current emissions, with similar results for nuclear energy (see SI, Table S12). Thus, provided enough carbon-free energy sources can be supplied, it is possible to create a near zero-emitting material system applying the co-recycling system proposed in this work. As for coal, gas and biomass as energy supply for the ATCR, these can result in both a reduction or an increase in the overall emissions.

Using biomass as an energy source could lead to emissions reductions when using pyrolysis, *Low* pathway, when accounting for the total emissions, including direct emissions from combustion as well as emissions from infrastructure and supply chain. When applying the gasification pathway, the total biomass emissions will be higher (113%) than today's, or only 19% if only infrastructure and supply chain emissions are considered. For the *High* pathway, Combustion with CCU, high amounts of electricity are required, increasing emissions to several times that of the base case, 680% in total). For that pathway, the infrastructure and supply chain emissions alone are higher than the base case synthetic C-materials emissions.

Using natural gas, the emissions could be reduced to 38% and 70% of the present emissions, for pyrolysis and gasification respectively, while if the gas is used to provide the electricity required for the combustion and CCU pathway, the emissions will double. Similarly, using coal for combustion and CCU will increase the emissions by about 5 times. Using coal to provide the pyrolysis heat will result in 52% of the base case emissions. For the gasification case, the emissions will remain similar to today.

## 4. Discussion

### 4.1. *Resource efficiency*

The base case shows that C-materials involve extensive use of resources and extensive losses throughout the supply chain. Assuming society will rely only on current recycling techniques, which have an efficacy of about 60%, and the totality of the 240MtC synthetic post-consumer waste was recirculated to manufacture new synthetic products, about 140MtC of synthetic products would be recycled. As a consequence, more than 60% of fossil resources will still be needed to cover the demand for synthetic C-materials. On the contrary, as shown

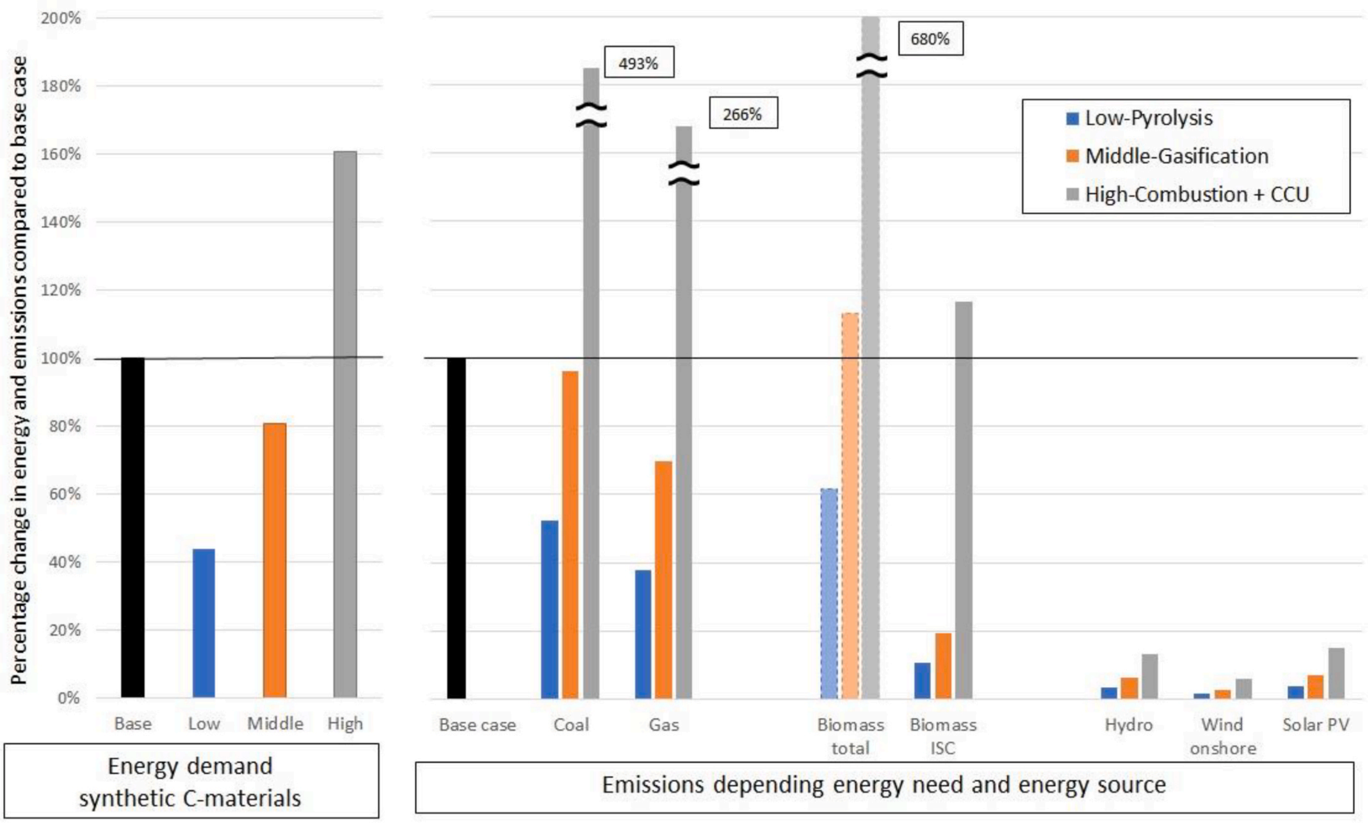

**Fig. 4.** Potential changes in energy demand (left) and carbon emissions (right), as compared to the base case. Biomass total includes direct and Infrastructure and supply chain emissions (ISC). Noted that impacts of the biomass direct net emissions are context-dependent, and biomass emits $CO_2$ that was previously sequestered from the atmosphere and will be sequestered again if biomass is sustainably sourced.

in the co-recycling scenario, the amount of carbon in post-consumer waste is sufficient to produce new C-materials via advanced thermochemical recycling has the potential to decouple the system from fossil resources. Indeed, the resource utilization ratio can improve from 0.7 to 1.1 $t_{C\text{-material}}/t_{CRtM}$, meaning that fewer carbon resources need to be extracted than C-material products are consumed. Similarly, the total losses of the CMS can be significantly reduced, resulting in a decrease in the resource losses ratio from 1 to 0.5 $t_{CRtM\text{-Losses}}/t_{C\text{-material}}$.

The co-recycling scenario shows a high potential for recirculation and use of post-consumer waste to produce new C-materials. Although the current amount of carbon in post-consumer waste is enough to close the synthetic C-material flows, C-materials use patterns can change over time. On the one hand, mechanical recycling and manufacturing process efficiencies will likely improve with technological developments. If so, these improvements will contribute to increasing the recirculation rate and reducing resource consumption. On the other hand, society may choose to use C-materials for longer or shorter timeframes. For instance, if the waste generation was reduced by half, an additional 235MtC will be needed for producing synthetic C-materials. Thus, other available flows should replace this deficit, such as the CRtM-Losses generated during the manufacture of natural C-materials. In such a case, while the losses ratio will decrease even further (0.3), the resource utilization ratio will remain the same. In contrast, if demand for C-materials increases and waste generation decreases additional carbon resources may be required, resulting in changes in the resource utilization ratio. It is noteworthy that when the patterns of product use change in a more wasteful society the extraction of new resources could still be avoided if there is an increasing demand for synthetic C-materials.

To compensate for changes in recycling efficiency and demand, other system losses or/and biomass can be used as a source of carbon. Plastics could also be mined from landfills or marine litter as a source of carbon to the CMS, potentially promoting the collection of already disposed plastics from landfills and oceans. Another alternative to achieve carbon recycling, not exploited in this work, is Direct Air Capture (DAC) of $CO_2$. The captured $CO_2$ can be converted back to plastics via synthesis and using hydrogen produced via electrolysis. However, as shown in the CMS analysis, there are enough carbon sources in today's material system that can be recovered with higher energy efficiency than applying DAC. Therefore, it can be assumed that DAC will only play a minor role in a future CMS if waste recirculation and losses reduction are prioritized.

Extracting carbon from existing waste deposits is in line with the circular economy, whereas introducing larger shares of biomass into the system would have the additional benefit of promoting negative $CO_2$ emissions. Indeed, a CMS that is fully decoupled from fossil fuels extraction behaves as a sink of biogenic carbon, or functional deposit, represented as stock in Fig. 3. This stock is about 190 MtC and 80 MtC, for natural and synthetic C-materials respectively. In the base case, the stock can be interpreted as long-term carbon storage that remains in the system in the form of products. Under the condition that biomass extraction has a neutral influence on land carbon stocks and sink strength, the proposed co-recycling scenario would have a functional deposit of 1 $GtCO_2$ net emissions.

### 4.2. Energy demand and carbon emissions

The energy demand of a co-recycling system that implements pyrolysis and/or gasification pathways can be lower than the energy demand of the base case (see Fig. 4), while a system relying solely on combustion and CCU implies an increased energy demand. Consequently, maximizing the share of pyrolysis in the portfolio of ATCR would be beneficial from an energy point of view. However, of the three technology pathways, direct recovery of monomers via pyrolysis is the most sensitive with regards to the quality of the feedstock, which means that the maximum share of waste that can be treated via pyrolysis is limited by the composition of the available waste.

Today, about 30% of carbon in the post-consumer waste generated are synthetic polymers that are prone to thermally decompose into monomers and other chemical building blocks via pyrolysis, i.e., PE, PP and PS (Scheirs and Kaminsky 2006). For the remaining 70% of the waste stream, the gasification and/or combustion with CCU route would be required for the recycling. Given the variety of waste compositions, including fractions of PE/PP/PS contaminated with other C-materials, a system based solely on the pyrolysis route is not realistic if the goal is to reach circularity. Circularity could, however, be achieved using the combustion with CCU pathway for all waste available, but this is at the expense of moving into a recycling system that has a higher energy demand than the base case. Therefore, a combination of the three ATCR technology pathways analyzed in this work should be necessary to achieve the double goal of circularity while keeping the energy demand below that of today.

In terms of carbon emissions, there is a potential to decrease the current emissions in the CMS by introducing different ATCR technology pathways as shown in Fig. 4. Generally, a co-recycling scheme involving ATCR reduces the emissions partly given that the embedded carbon in waste is not emitted, but also since the pyrolysis and gasification pathways require less energy than the base case. Yet, the benefits in terms of emissions of introducing ATCR is highly dependent on the energy mix of the contextual *Energy System*. The results show that energy-related emissions have a large impact on the total carbon emissions, and that renewable and low carbon electricity is paramount to justify a transition to ATCR recycling from a carbon emissions standpoint.

For regions with high availability of renewable electricity from hydro, wind and solar, C-materials could be recycled by the CCU route. For wind and hydropower systems the emission reduction will be more than 95% compared with today's system, even when using the CCU pathway, which, as mentioned above, is more energy-intensive than via pyrolysis and gasification. In order words, the CCU pathway only seems sensible for regions with high availability of low-cost renewable electricity. Under these conditions, a socio-economic optimization may lead to a pure CCU recycling system.

In regions with coal, gas or biomass dominating the energy mix, the initial pathway to create circular C-material flows will be via the pyrolysis and gasification pathways, which will reduce the energy demand, but not offer a significant carbon emissions reduction potential. If countries can overcome the challenge of modifying their electricity system towards zero carbon emissions, the process can be electrified reducing the need for supplementary combustion (of coal, gas or biomass) for process heat. In this case, it is possible to switch from coal/gas or biomass combustion to waste combustion followed by CCU, including hydrogen production via electrolysis. Therefore, to avoid lock-in effects, the synthesis downstream of the gasifier should be flexible and be adapted to predominantly use $CO_2$ and $H_2$ instead of CO and $H_2$. This will influence the operation, but if this development is already accounted for in the design of the plant, the transition from carbon fuels to electricity to cover the energy demand can be made without a costly redesign of the plant.

In addition, when using coal, gas or biomass, if the $CO_2$ stream is concentrated, it allows for $CO_2$ capture followed by geological storage (CCS). This would reduce the climate impact, with a slight increase in energy demand and with the possibility to create negative emissions when using biomass. Nevertheless, it should be noted that impacts of the biomass use on net emissions are context-dependent and influenced by the feedstock, management regime, soil and climate conditions, the scale of deployment, land use and supply chain and infrastructure emissions (Cowie et al., 2021). Biomass systems can have anywhere from a large beneficial mitigation effect, if the biomass use results in large net emissions savings and land carbon increases, to an undesirable effect when land carbon losses outweigh any emissions savings for a relatively long time period (IPCC et al., 2019b; 2019c). Deployment strategies for biomass, bioenergy and bioenergy with CCS need to encourage appropriate biomass production and supply chains. Such strategies include the protection of ecosystems of high ecologic value and/or with large land carbon stocks, and incentivizing land-use management to protect land carbon stocks and environmental functions while increasing land productivity (Amelung et al., 2020; Ittersum et al., 2013).

### 4.3. Towards a circular economy

As shown, there is the potential for recirculation of different flows to produce new C-Products via advanced thermochemical recycling, where these ATCR technologies are the key to enabling 100% carbon recovery. While pyrolysis and gasification have been proven under demonstration scale for homogenous waste streams (Silveira, 2020). There is a need for technical development to make pyrolysis and gasification work with heterogeneous wastes in a continuously and robust way (Kusenberg et al., 2022; Quicker et al., 2022) as well as effective carbon capture technologies (Kähler et al., 2021; Thunman et al., 2019). However, if society decides on the right policy measures to promote circularity it can be argued that it is not the technology that limits the recycling proposed in this work. Instead, the collection and recirculation of all the flows may be challenging in practice, and there may be a long transition period until high waste collection rates are achieved globally.

High collection rates are achieved in high-income countries, but the collection rates are as low as 38% in low-income countries (Kaza et al., 2018). In addition, some flows can be difficult or expensive to recover, thus, they may be a long transition period leading up to 100% collection being achieved on a global scale. Yet, the application of policy measures – e.g., a phase-out of allowing plastics to be produced from fossil oil - which fosters the deployment of a co-recycling scenario will create a value for the waste, in particular, plastic waste, which may incentivize collection. Effective waste collection systems are paramount for a global co-recycling system to be feasible. However, even when an extensive waste collection system is in place, the leakage-prone C-materials (e.g., littering and mismanagement) can be hard to stop. In those cases, behavioural changes (Dunstan, 2022) and high infrastructure costs may be required (for instance filters in each building to avoid textile washing microplastic to end up in the environment or traps in each river to avoid litter to end up in nature). Therefore, increasing recirculation and recycling does not mean an end to littering. For these leakage-prone products, design changes, a ban or biodegradable alternatives may be the way forward. The design of future material systems and the waste handling should consider the trade-off between the production of biodegradable products and maintaining the value of the Carbon molecule which can be repeatedly recirculated.

A co-recycling system with ATCR enables the use of any form of carbon, allowing the recovery of carbon that cannot be recovered using traditional methods, such as the carbon from contaminated waste, plastic litter, and producing high-quality synthetic C-materials. Attaching a value to the carbon molecule itself may incentive collection and accounting for the carbon embodied and within the supply chain would alleviate the environmental burdens related to carbon emissions and deposition. However, it must be noted that other elements are also present in C-material waste, such as sulfur, chlorine, or metals. In a real system, taking care of the emissions of those compounds and other possible environmental burdens needs to be considered, similar to what is already done in waste to energy facilities. Thus, circularity indicators, other than carbon, would be necessary for a complete CE (Eriksson, 2022; Papadopoulos, 2020). Yet, a possible benefit of applying ATCR is that, implemented at a large scale, it opens the possibility to develop circularity of other minor components.

For a circular economy of C-materials, not only there is the need to implement a combination of advanced thermochemical recycling technologies, but also to implement waste collection systems worldwide. In addition, for deep decarbonization, the availability of renewable energy is paramount. Thus, economic incentives and policies which phase out the extraction of fossil resources for material production are required. The optimal solution depends on the socio-economic regional conditions, which depend on the region's economic prosperity and geographical conditions for local energy sources, as well as the development of the waste management system.

## 5. Conclusion

A co-recycling scenario utilizing advanced thermochemical recycling is proposed and analyzed. The results show that such a scenario can combine natural and synthetic C-materials in the same recycling scheme and that enables to decoupling of the Carbon Material System from the extraction of fossil resources, as well as minimizing waste generation. Advanced thermochemical recycling enables the recovery of any carbon flow ($CO_2$, natural or synthetic, losses, etc.) and the production of high-quality products. A focus on carbon recycling, instead of material recycling, can improve C-materials (wood, paper, plastics, and textiles) resource management, by increasing the resource utilization from 0.7 to 1.1 $t_{C\text{-material}}/t_{CRtM}$, i.e., producing more C-materials than resources used, as well as reducing the CMS losses by half.

Global application of advanced thermochemical recycling technologies has great potential: less energy than used in today's material system may likely be required, and carbon emissions can be reduced using different energy sources, leading to near-zero carbon emissions with renewable energy. This work gives a first insight into the possibility of closing the carbon flows within the Carbon Material System and the energy and emissions improvements that are possible, depending on technology choice, waste management conditions and energy mix. The co-recycling system could benefit the full C-material value chain, preserving the value of resources and maintaining the stock of materials in society, and reducing losses. However, further work is needed to identify deployment strategies and to explore further the integrated technologies, as well as the energy and economic implications of deployment.

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## Data and materials availability

All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Information. Additional data related to this paper may be requested from the authors.

## CRediT authorship contribution statement

**Isabel Cañete Vela:** Methodology, Validation, Formal analysis, Investigation, Data Curation, Writing – Original Draft, Visualization. **Teresa Berdugo Vilches:** Methodology, Validation, Writing – review & editing. **Göran Berndes:** Validation, Writing – review & editing. **Filip Johnsson:** Validation, Writing – review & editing. **Henrik Thunman:** Conceptualization, Methodology, Validation, Writing – review & editing.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

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