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Time-integrated GHG emissions in advanced waste-to-energy plants producing fuels, chemicals and electricity from MSW refuse

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THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

Time-integrated GHG emissions in advanced waste-to-energy plants producing fuels, chemicals and electricity from MSW refuse

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Preface

During my studies in Environmental Science, I thought my future was the protection of the environment; I imagined a future in direct contact with the Nature since that seemed to be the best way to protect it. Right after I finished my bachelor degree, I understood that the protection starts much before I had thought, i.e., before the pollution and the damage. Thus, the prevention has become my tool to avoid the impact of human activities to the environment. I became an Environmental Inspector and Auditor; and I learned very much about environmental regulation and, particularly, about waste management and so I realized my future was to contribute to the prevention and minimization of the environmental impacts in the management of municipal solid waste (MSW).

A postgraduate in Renewable Resources and Environmental Engineering gave me the technical background for this doctoral thesis. During the doctoral training, my life has been a fight for developing a tool able to calculate time-integrated GHG emissions in the use of MSW refuse for the production of a mix of products and services in advanced waste-to-energy (WtE) plants. This report is the result of that effort, hard but rewarding, where hard work and sacrifice are mixed with the pride of a job well done and the luck to have meet great friends and great experts.

Appended papers

I. GHG saving in thermochemical biorefineries using municipal solid waste in Andalusia
Aracil C, Haro P, Ollero P, Vidal-Barrero F (2014)
Proceedings of the *22nd European Biomass Conference and Exhibition, EUBCE* 2014, Hamburg, Germany, pp 1576 - 1578

II. Time-integrated GHG emissions in a thermochemical biorefinery producing ethanol from municipal solid waste in Andalusia
Aracil C, Haro P, Ollero P (2015)
Proceedings of the *23rd European Biomass Conference and Exhibition, EUBCE* 2015, Vienna, Austria, pp 1376 – 1379

III. Proving the climate benefit in the production of biofuels from municipal solid waste refuse in Europe.
Aracil C, Haro P, Giuntoli J, Ollero P (2017)
Journal of Cleaner Production. 142: 2887–2900

IV. Contrasting the greenhouse gas removal potential of carbon capture and storage and renewable-derived plastics in advanced WtE plants
Aracil C, Giuntoli J, Cristobal J, Haro P (2017)
Submitted for publication to the International Journal of Greenhouse Gas Control (under review)

V. Implementation of waste-to-energy options in landfill-dominated countries: Economic evaluation and GHG impact
Aracil C, Haro P, Fuentes-Cano D, Gómez-Barea A (2017)
Manuscript for the journal Waste Management

VI. Dynamic assessment of Waste-to-Energy schemes in current European landfill-dominated regions
Aracil C, Haro P, Fuentes-Cano D, Gómez-Barea A (2017)
Proceedings of the 25th European Biomass Conference and Exhibition, EUBCE 2017, Stockholm, Sweden, 12-15 June, 2017

Co-authorship statement

In all papers, Cristina Aracil has been the main author and carried out the definitions of the systems and scenarios, calculations, participation in the discussion, writing of the original manuscript and preparation of most figures. Pedro Haro has defined the structure of the papers, supervising them all except Paper V, which was supervised by Alberto Gómez-Barea with the assistance of Pedro Haro. Pedro Ollero supported the discussions in Papers I-III. Jacopo Giuntoli collaborated in the definition of the systems and the methodology for Papers III and IV and participated in the discussion and writing. Jorge Cristobal collaborated in the discussion and writing of Paper IV. Fernando Vidal-Barrero and Diego Fuentes-Cano participated in the discussion of Papers I and V-VI, respectively.

Related papers that are not included in this thesis

- Balance and saving of GHG emissions in thermochemical biorefineries.
  Haro P, Aracil C, Vidal-Barrero F, Ollero P. (2015). *Applied Energy*; 147:444-55.
- Rewarding of extra-avoided GHG emissions in thermochemical biorefineries incorporating Bio-CCS.
  Haro P, Aracil C, Vidal-Barrero F, Ollero P. (2015). *Applied Energy*; 157:255-266.

These papers where part of the initial research training of Cristina Aracil in the early stages of her doctoral studies.

TABLE OF CONTENTS

A mi hija Sofía

Abstract

The evaluation of the climate benefit in the production of fuels from conventional biomass has recently evolved by incorporating a dynamic approach, a comparison with the reference system and other recommendations from the IPCC. Important drawbacks have been identified in the comparison of conventional (static) and dynamic (time-integrated) assessments for the production of biofuels. This thesis contributes to a better understanding of the real climate benefit in the production of products and services using a specific and plentiful waste in Europe, i.e., MSW refuse (the unsorted stream of MSW usually disposed of). Lately, some dynamic assessments have been made for the production of fuels and electricity using forest and agricultural residues. In this thesis, the existing work is expanded by considering a residue (MSW refuse) which is already in-use within the different regional waste management schemes in Europe (incineration and landfilling) and the production of a material that stores biogenic carbon, i.e., renewable-derived plastic materials. The climate benefit of the proposed advanced waste-to-energy (WtE) plant is evaluated by defining two systems (the one proposed in the thesis and the reference system) using system expansion and substitution. The dynamic modeling of the waste management scheme in Europe (current and future) as well as the temporary storage of the biogenic carbon fraction in the renewable-derived plastics (intimately related to the management scheme) are the main contributions to the field. The proposed methodology is based on two climate benefit indicators: the climate mitigation index (CMI) and the differential climate impact (DCI). The indicators analyze the impact of the replacement of the current waste management system for one based on advanced WtE plants. In Paper I, a preliminary work applying the static methodology (GHG balance) is carried out for the analysis of the results in advanced WtE plants producing biofuels, drop-in chemicals and electricity and with the possibility of carbon capture and storage in bioenergy (Bio-CCS). In Paper II, the dynamic GHG emission assessment is applied to the advanced WtE plant analyzed in Paper I. In Paper III and IV, two countries are selected for the comparison of the systems: Spain, where landfilling is dominant; and Sweden, where incineration is dominant. Moreover, two different scenarios are taken into account: Scenario 1, in which the reference system remains unaltered, and Scenario 2, in which there is an evolution towards landfill banning and decarbonization of the energy mix. The results reveal that the landfilling replacement in dominant-landfill European countries has a positive climate impact in the short term, although the long-term impact depends on the evolution of the reference system (waste management and electric mix). Renewable-derived plastics are proposed (Paper IV) as an alternative greenhouse gas removal (GGR) technology and compared with Bio-CCS as the common GGR technology in most Integrated Assessment Models (IAMs). The production of plastics compares 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. In Paper V and VI, the static and dynamic assessment for the production of electricity is analyzed.

Resumen en castellano

En esta tesis se desarrolla el análisis dinámico (integrado en el tiempo) de las emisiones de gases de efecto invernadero (GEI) asociadas a la producción de productos y servicios en un planta de gasificación (tratamiento avanzado de valorización energética) alimentada por la fracción rechazo de los residuos sólidos urbanos (RSU), es decir la fracción no reciclable ni compostable que va a depósito en vertedero o incineración. Tradicionalmente, el cálculo de la huella de carbono de un proceso alimentado por fuentes renovables se ha llevado a cabo desde un punto de vista estático, mediante métodos exhaustivos que permiten calcular las emisiones de GEI asociadas al ciclo de vida completo de un proceso o producto y cuyo objetivo es la comparación de alternativas para producir una misma unidad funcional (por ejemplo, una misma planta de producción que puede ser alimentada por diferentes tipos de biomasa) incluyendo la alternativa de origen fósil. Sin embargo, el resultado final de estos estudios estáticos es un valor promedio de las emisiones de GEI anuales, un resultado que puede ser insuficiente cuando la fuente renovable que se analiza es un residuo que forma parte de un ciclo biológico de degradación que se usa como sistema de referencia para valorar si ese residuo debe ser utilizado como fuente energética o no. Por otro lado, recientemente se están llevando a cabo estudios dinámicos de emisiones centrados en residuos agrícolas y forestales; sin embargo, la fracción rechazo de los RSU tiene un potencial energético similar a los residuos agrícolas en Europa. Los estudios dinámicos analizan la evolución de las emisiones de GEI producidas en un proceso de degradación de la materia (normalmente residuos) y los comparan con un sistema de referencia siguiendo las recomendaciones del Panel Intergubernamental por el Cambio Climático (IPCC por sus siglas en inglés). La fracción rechazo de los RSU puede proceder bien de plantas de tratamiento mecánico y biológico (TMB) o de la separación en origen, siendo la práctica más común en Europa su depósito en vertedero (especialmente en las regiones del sur de Europa). El depósito en vertedero produce emisiones de GEI que empiezan unos pocos meses o incluso años después del depósito y continúan hasta 40 años después con diferente intensidad a lo largo del tiempo. De ahí la necesidad de aplicar un enfoque dinámico a un estudio donde el sistema de referencia es el vertedero. Esta tesis estudia, en clave del potencial beneficio climático, la sustitución del sistema actual de gestión de la fracción rechazo de los RSU (vertedero e incineración) por un sistema basado en plantas de gasificación avanzadas. Estas plantas producirían un mix de productos (combustibles y/o químicos de origen renovable) y/o servicios (electricidad y/o calor). Se analizan dos familias de plantas de gasificación avanzada: por un lado, grandes plantas de gasificación llamadas biorrefinerías termoquímicas que producen combustibles/químicos así como electricidad/calor con una alta eficiencia energética. Por otro lado, plantas de gasificación de pequeña y mediana escala produciendo electricidad. Dado que las plantas de producción de electricidad (pequeña/mediana escala) son susceptibles de una implementación a corto plazo, también son analizadas desde el punto de vista económico.

Los plásticos de origen renovable (fabricados a partir de los químicos generados en la biorrefinería termoquímica) son propuestos como una tecnología alternativa a la captura y almacenamiento de carbono en bioenergía (Bio-CAC) para la retirada de carbono de la atmósfera. En el caso de los plásticos renovables el carbono queda retenido en el plástico durante un periodo de tiempo que dependerá del tipo de plástico, su tiempo de vida y el sistema convencional de gestión de los residuos plásticos. La posibilidad de incorporar alternativamente o en conjunto el Bio-CAC (tecnología comúnmente analizada en los Modelos de Evaluación Integrados, IAMs en inglés) y los plásticos de origen renovable se analiza en detalle. Los resultados revelan que la producción de plásticos de origen renovable es comparable en términos de beneficio climático a corto y medio plazo e incluso podría suponer un mayor beneficio climático a largo plazo en países donde la incineración es una práctica predominante (típicamente en el norte de Europa).

Desde el punto de vista metodológico, dos países han sido analizados: España, que representa a los países del sur de Europa donde el depósito en vertedero es predominante y Suecia, representando a los países del norte de Europa donde la incineración es predominante. Para el análisis, dos posibles escenarios son considerados, un escenario 1 donde el sistema de gestión de residuos y el mix energético no varían a lo largo del tiempo (este escenario permite la comparación con una evaluación estática de emisiones desarrollada también en esta tesis) y un escenario 2 que evoluciona hacia un sistema sin depósito en vertedero y un mix energético (electricidad) bajo en carbono. En ambos escenarios se estudia un horizonte de tiempo de 100 años según recomienda el IPCC.

De la colaboración con el Joint Research Centre (JRC, Comisión Europea) surgió una propuesta metodológica basada en dos nuevos indicadores de beneficio climático: el índice de mitigación climática (CMI, en inglés) y el diferencial de impacto climático (DCI, en inglés). Del mismo modo, el JRC colaboró en el modelado del carbono biogénico almacenado en los plásticos de origen renovable que dio lugar a un parámetro para ser incorporado en un análisis estático de emisiones ($\bar{e}_{pool,}$ promedio de carbono biogénico almacenado en plásticos de origen renovable) y en uno dinámico ($e_{pool,}$ sumatorio de las corrientes de reciclaje, vertedero e incineración a lo largo del tiempo). Por tanto, el modelado dinámico del sistema de gestión de residuos (actual y futuro) así como el modelado del almacenamiento de carbono biogénico en los plásticos de origen renovable son las principales contribuciones de esta tesis.

Los resultados revelan que un análisis estático de las emisiones temporales no es suficiente para la toma de decisiones sobre la sustitución del vertedero por plantas de gasificación avanzadas. El impacto de esta sustitución tiene un efecto positivo a corto plazo para ambos tipos de plantas aunque el concepto de biorrefinería termoquímica produciendo plásticos de origen renovable cursa con el mayor beneficio climático. El beneficio a largo plazo no estaría garantizado a no ser que se tomen medidas para la restricción del depósito en vertedero.

1. Introduction

1.1. Residual biomass and MSW refuse

According to the Renewable Energy Directive (RED) (Directive 2009/28/EC[1]), 'biomass' means the biodegradable fraction of products, waste and residues from biological origin, from agriculture (including vegetal and animal substances), forestry and related industries including fisheries and aquaculture, as well as the biodegradable fraction of industrial and municipal waste. In the last decades, biomass has become a promising alternative to fossil fuels in the production of energy. However, non-residual biomass has associated upstream greenhouse gases (GHG) emissions (cultivation, harvesting, direct and indirect land-use change) that could compromise the sustainability of the process (Directive 2015/1513/EC, Lapola et al., 2010, Gerssen-Gondelach, et al., 2016). For instance, in the case of the energy crops, one of the first impacts is the displacement of food crops needed for human and animal feeding (direct land-use change, LUC) and other is the need of finding new land (maybe not as productive as the first) where set up the displaced food crop (indirect land-use change, ILUC). A higher use of the land involves a higher climate impact over it e.g. a higher degradation and loss of biodiversity; and this impact can be difficult to estimate due to the uncertainties associated to ILUC e.g. agriculture and food security or others land uses (Directive 2015/1513/EC, ILUC Quantification Study of EU Biofuels).

On the contrary, residual biomass has not upstream GHG emissions associated. According to the previous definition of biomass, the biodegradable fraction of municipal solid waste (biowaste) is considered as biomass. In fact, biowaste is currently used in the production of compost or in the production of biogas by anaerobic digestion (energy valorization). Both options fulfill the waste hierarchy[2] prioritizing first the recycling and then the valorization before the disposal (Directive 2008/98/EC) and therefore it would seem that there is no need to change the current situation. However, the waste hierarchy is not the unique tool to make decisions about waste management. The European regulation establishes targets to be achieved in order to reduce the land disposal in Europe (Directive 2008/98/EC). Because of that, it is necessary to find alternatives to manage the huge amount of wastes going to landfill disposal in Southern and Eastern Europe e.g. in Spain the 55% of the municipal solid waste (MSW) generated is landfilled every year; whereas in

[1] Directive 2009/28/EC was further amended by Directive 2015/1513/EC but the concept of biomass and the aspects affecting this thesis, except those specified in the text, remain in force in the original Directive.
[2] http://ec.europa.eu/environment/waste/framework/

the Northern Europe, landfilling ratio is below incineration and recycling ratios being even negligible in some countries e.g. zero landfilling in Germany (Figure 1).

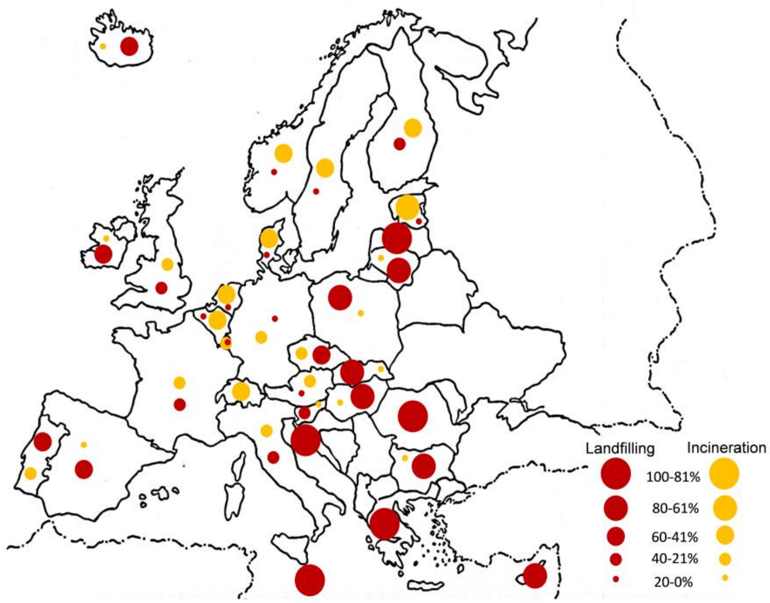


Figure 1. Landfilling and incineration ratios regarding total MSW generated in Europe (Aracil et al., 2017).

MSW refuse is the unsorted stream of MSW going currently to landfill disposal or incineration. This stream has two possible origins depending of the waste collection method. The first one is the stream not separated at source and collected in a communal bin. The second one, it is the refuse of mechanical biological treatment (MBT) plants fed by a mix of wastes and where the compostable and recyclable fraction has been separated. Therefore, MSW refuse is a heterogeneous mix of waste streams whose sorting is not viable. This stream is partially biodegradable; hence, it is not considered as pure biomass. However, MSW refuse usually contains a biodegradable fraction over 50% (IEA, 2012). Considering a lower heating value around 9 MJ/kg (Boesch et al. 2014, Consonni and Viganò et al., 2012 and Yassin et al., 2009), the MSW refuse generated in Europe would be equivalent to 1,250 PJ/year. Therefore, MSW refuse in Europe has a similar potential to agricultural residues, which are already considered to play an important role in the future bioeconomy (Philippidis et al., 2016).

On the other hand, the urgent need of reducing GHG emissions in order to limit the global warming makes the search of alternatives to the use of fossil fuels essential (Directive 2009/28/EC). Moreover, waste disposal (landfilling or mass-burnt incineration) should be replaced by waste-to-resource alternatives in order to fulfill the objectives of the Circular Economy (Report from the Commission to the European Parliament, 2017). Therefore, an advanced waste-to-energy (WtE) plant using MSW refuse as a feedstock for the production of products (biofuels and/or renewable-derived plastics) and/or services (electricity and/or heat) by means of gasification would be an interesting system for the reduction of GHG emissions.

1.2. Situation of landfill disposal as a part of the MSW management in the world, Europe and Spain

Land disposal is still by far the most popular option among the waste management options in the world since its cost is low, e.g. the gate fee, the charge levied to a quantity of waste received at a waste processing facility, is lower for a landfill than for a WtE plant (CEWEP, 2016). In Figure 2, MSW treatment ratios are shown for Europe and some more selected countries (Eurostat, 2014, EPA, 2014, Mian et al., 2016). The landfilling ratio for Spain, Romania, USA or China is over 50% and two reasons for this ratio can be considered: on the one hand, the level of development, e.g. Romania is one of the poorest European countries; and, on the other hand, the land availability, e.g. USA is between the five largest countries in the world. Singapore is a clear example of country with land scarcity and this country just landfills the 3% of their MSW generated.

Hence, landfilling requires the use of a not always available land and it has associated several environmental impacts over land, atmosphere, hydrosphere and biosphere. Because of that, the most developed countries are evolving towards alternatives to manage their waste being energy recovery the most usual alternative to landfilling.

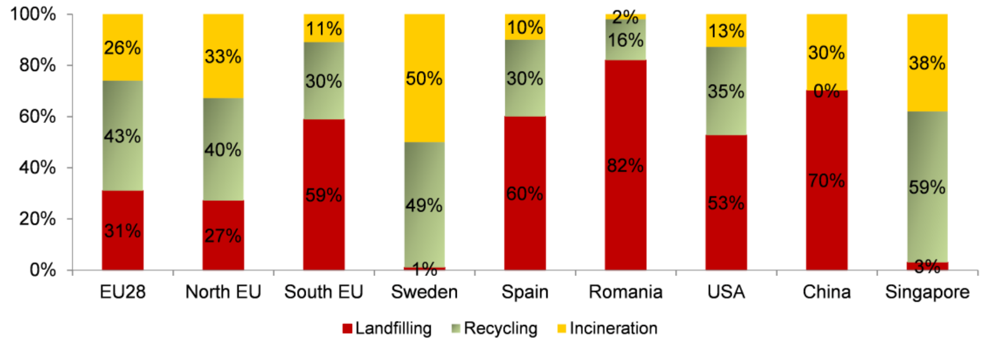


Figure 2. MSW treatment (landfilling, recycling and incineration) ratios regarding total MSW generated (mass basis) in Europe, USA, China and Singapore (Eurostat, 2014, EPA, 2014, Mian et al., 2016).

In Southern and Eastern Europe, landfilling involves a variety of dumping and landfill sites of different volumes where waste is disposed to different depths using different covers and different degasification methods. The environmental impact of the landfilling depends on their characteristics and the state of its management, e.g. landfill emissions are directly proportional to the landfill site volume and inversely proportional to the biogas fraction collected and burned. Opened-air dumping sites are still common in many Eastern European countries, e.g. Serbia or Romania, where a first task would be the transition towards modern landfills where waste is pretreated, buried and further degasified (Stanisavljevic et al., 2012). In fact, recent studies determine the methane concentration in the atmosphere is rising dramatically in the last decades. This methane is released from different sources but two thirds of the emissions are attributable to anthropogenic activities related to agriculture and waste management (Saunois et al., 2016, Global Carbon Project, 2001). In the waste sector, landfilling represents an important source of methane emissions. Because of that, mitigation strategies, e.g. covering and degasifying landfills, are determinant to reduce CH_4 emissions (Saunois et al., 2016, Global Carbon Project, 2001). However, dominant-landfill European countries should also encourage WtE initiatives in order to minimize GHG emissions and approach EU legislation (Nikolic et al., 2017).

In December of 2015, the first draft on Best Available Techniques (BAT) Reference Document for waste treatment was published (BAT, 2015). However, the European

Federation of Waste Management and Environmental Services (FEAD) has highlighted the limitations of this document to represent the waste management in Southern and Eastern Europe and the lack of recycling promotion in favor of landfilling. In order to face the challenge of minimizing the landfill impact, several projects have been carried out in Europe as for example the LIFE project called *Good practice on energy and products from biomass and waste* that finances projects as *Assessing, Capturing & Utilising Methane from Expired and Non-operational landfill* whose aim is to demonstrate techniques for valorizing and mitigating methane emissions from closed landfill sites (ACUMEN).

In Spain, recent news have commented the problem associated to waste management. These news are related to different issues but landfilling is one of the most usual topics. For instance, landfill restriction in Northern Europe entailed the promotion of WtE plants and that has led Nordic countries as Norway and Sweden into a struggle to import wastes from other European countries to feed their plants (2016/10). The environment risks of illegal waste treatment plants (2016/05 and 2016/08) and the growing role of methane (from landfilling) in anthropogenic climate change (2016/12) are also current issues in Spain (El País, 2016). Therefore, waste management is nowadays a social, environmental and techno-economic challenge in the world since huge amounts of wastes and residues are produced every year and their management is country-dependent.

Landfilling produces biogas composed mainly by CO_2 and CH_4. This biogas is produced by the partial degradation of the biodegradable fraction; i.e. a part of the biodegradable fraction remains unaltered and stored in the landfill site. Therefore, this biogas is completely biogenic. However, methane contributes twenty-five times more than dioxide carbon to the global warming (IPCC, 2014). Because of that, methane emissions from landfilling must be accounted and considered as environmental burden (Figure 3). As it was commented before, biogas recovery is not common in all the dominant-landfill countries and, when it is incorporated, only a part of the biogas is captured and combusted to generate electricity or burned in flare. Hence, emissions are released in the electricity production process (biogas combustion) and biogas leaks from the landfill site. Biogas leaks are not constant in the time, they represent a slow and uneven process with different intensity over time, and therefore, the temporal aspect of these emissions must be taken into account. In this thesis, a dynamic GHG emission assessment has been proposed and developed in order to integrate the time in the GHG emission calculation.

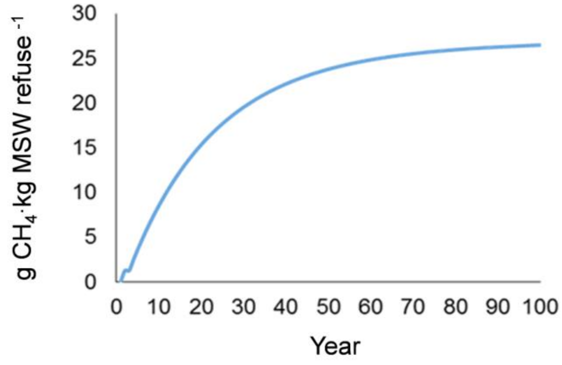


Figure 3. Methane emission profile of a landfill in Southern Europe (calculated from IPCC, 2006).

1.3. Conventional and Advanced WtE plants

The conventional WtE plant is based on incineration, which consists on the complete combustion of the MSW refuse to produce heat that is used directly or converted into electricity in a power plant. The most common technology used for incineration is a moving grate allowing the use of MSW refuse without pretreatment. In Southern Europe, electricity is the main product in combined heat and power (CHP) plant, since heat demand is generally low, so that high electrical efficiency is required. To produce electricity with reasonable efficiency (>20%) large scale is necessary (>0.5 Gt RDF/y) (Consonni and Viganò, 2012). Therefore, MSW refuse is transported from several production sites, e.g. mechanical and biological treatment (MBT) plants or transfer stations to a centralized large-scale incineration plant.

The advanced WtE plant is based on gasification of RDF to produce a syngas, which in the context of the present study is further combusted to produce electricity/heat or converted into fuels/drop-in chemicals (for the production of renewable-derived plastics). Gasification technologies can be adapted to the volume of waste available (medium scale, i.e. lower than 100 kt/y) to produce electricity (Arena et al., 2015). Several technologies can be considered for the production of syngas (moving grate and fluidized bed gasifiers). Fluidized bed gasification (FBG) technologies require a homogeneous fuel, so that the MSW refuse has to be pretreated and converted into refuse derived fuel (RDF). Moving-grate gasifiers can generally be operated with direct MSW refuse but the lower technical development of gasification compared to incineration suggests to pretreat the refuse in order to improve the availability of the gasification process and the increase in the heating value of the feedstock (Table 1)

Table 1. Technologies for conventional and advanced WtE plants of interest in this thesis.

	Technology	Scale	Feedstock
Conventional WtE	Moving grate combustor (GC) and steam Rankine cycle (SRC)	Large	MSW refuse
Advanced WtE	Fluidised bed gasifier (FBG) and internal combustion engine (ICE)	Medium-small	RDF from MSW refuse
	FBG and organic Rankine cycle (ORC)		
	Moving grate gasifier (GG) and SRC		

1.4. Static vs. Dynamic GHG emission assessment for MSW refuse. The role of dynamic GHG emission assessment in the evaluation of greenhouse gas removal technologies

Several tools have been developed in the literature for the calculation of GHG emissions of processes using biomass (Guinée et al., 2009, Yang and Zhang, 2011). In this thesis, the GHG balance is defined as an annual average of all anthropogenic cradle-to-grave GHG emissions in the production of products and/or services using MSW refuse. GHG balance can be seen as a simplified version of a carbon footprint assessment[3], which aims to be useful for the calculation of a GHG saving of a process (biofuel production) or it is applied to new process concepts still far to be commercial (advanced WtE plants).

[3] The conventional environmental assessment (carbon footprint assessment) would be carried out using a Life Cycle Assessment (LCA) limited to the GHG emissions of the whole process (cradle-to-grave). LCA is a well-known tool, which analyzes all relevant emissions and resources consumed and the related environmental and health impacts and resource depletion issues that are associated with any goods or services (EUR 24708 EN, 2010). The carbon footprint assessment is a scientific tool useful for the comparison of new products and technologies, which requires of precise data (inventory) and well-defined system boundaries. Therefore, it is necessary to have a detailed knowledge of the full life cycle of the product (from the extraction of resources, through production, use, and recycling, up to the disposal of remaining waste (EUR 24708 EN, 2010)) and the fossil reference (the LCA is in essence a comparison of different alternatives to produce the same functional product). LCA requires a relevant functional unit and well-defined system boundaries. The GHG balance uses MJ of products as a functional unit, therefore, it would not be a consistent method compared to LCA standards.

The GHG balance allows a static assessment of the GHG emissions where the value achieved is an average of the annual GHG emissions, i.e. the emissions are supposed to be constant along time. However, static GHG assessments are a simplification of the actual GHG emissions associated to the process. This simplification cannot be enough for dynamic processes. For instance, degradation is a time-dependent process, so the assessment of the moment and the duration of the emissions is essential. In the case of agricultural and forest residues for bioenergy production, the usual alternative is to leave them on the ground degrading and incorporating organic carbon to the soil (Giuntoli et al., 2015b, Gaudreault et al., 2015, Guest et al., 2013b, Gustavsson et al., 2017). This degradation is a slow and uneven process generating emissions with different intensity over time (similar to landfilling). Time factor has to be taken into account to decide if it is better to use the residues as feedstock or to leave on the ground. In the case of MSW refuse, the timing of GHG emissions in the landfill has to be modeled. Therefore, a dynamic GHG assessment is supposed to be necessary in studies about emissions from the residues degradation.

Moreover, dynamic GHG assessment allows studying the evolution and/or changes in a process and their impact on the climate. For instance, the waste management scheme in Europe is expected to change along time in order to fulfill the environmental regulation, it is a scheme subject to a slow but constant evolution. This evolution would be only an average in a static GHG assessment. On the other hand, dynamic GHG emission assessment is an efficient tool to assess the potential climate impact of greenhouse gas removal (GGR) technologies. GGR technologies (Table 2) are a type of climate engineering aiming to remove greenhouse gases (GHG) from the atmosphere, and thus tackling the root cause of global warming. These techniques either directly remove GHG, typically CO_2, or alternatively seek to influence natural processes to remove GHG indirectly.

Table 2. Summary of GGR technologies.

Afforestation and reforestation
Wetland restoration
Agricultural soil sequestration
Biochar
Bioenergy with carbon capture and storage (Bio-CCS)
Renewable-derived plastics[a]

[a] Proposed in this thesis.

Bio-CCS allows capturing the biogenic CO_2 produced in industrial processes using biomass or wastes and avoiding its emission to the atmosphere, yielding a net removal of CO_2 from the atmosphere (Koornneef et al., 2012). The production of renewable-derived plastics is proposed in this thesis as an alternative GGR technology to Bio-CCS. Renewable-derived plastics are plastics made from biomass (also bio-plastics[4]) or wastes that represent a biogenic carbon storage in stable chemical structures. The storage period for a renewable-derived plastic depends on the type of plastic, on the lifetime and on the treatment applied to the plastic waste at the end of its lifetime (Guest et al., 2013a, Wang et al., 2014, Levasseur et al., 2013, Pawelzik et al., 2013, Vogtländer et al., 2014). Recycling of waste plastics becomes crucial to extend plastics lifetime, reducing the consumption of fossil fuels and the environmental footprint. Depending on the plastic material, there is a maximum number of recycling cycles and therefore a limit to their overall lifetime. However, not all plastic materials are recycled and each recycling cycle generates a waste refuse that cannot be further recycled. Hence, every year around 70% of post-consumer plastics waste is either landfilled or incinerated (with energy recovery) in Europe (Plastics Europe, 2015).

In the carbon cycle, the carbon previously captured by photosynthesis (1) comes back to the atmosphere by means of breathing and combustion processes (2) but a fraction of the carbon is stored permanently, e.g. fossil reservoir and long-lived trees (3), or temporally, e.g. living beings and soil (4). In the Bio-CCS process, an extra fraction of carbon is stored permanently in a geological reservoir (5). In the case of the renewable-derived plastics, an

[4] Renewable-derived plastics is a term preferred to bio-plastic in this thesis since MSW refuse is not pure biomass.

extra fraction of carbon is stored temporally (6), e.g. plastics in use in the market, or permanently, e.g. plastics landfilled (7) (Figure 4).

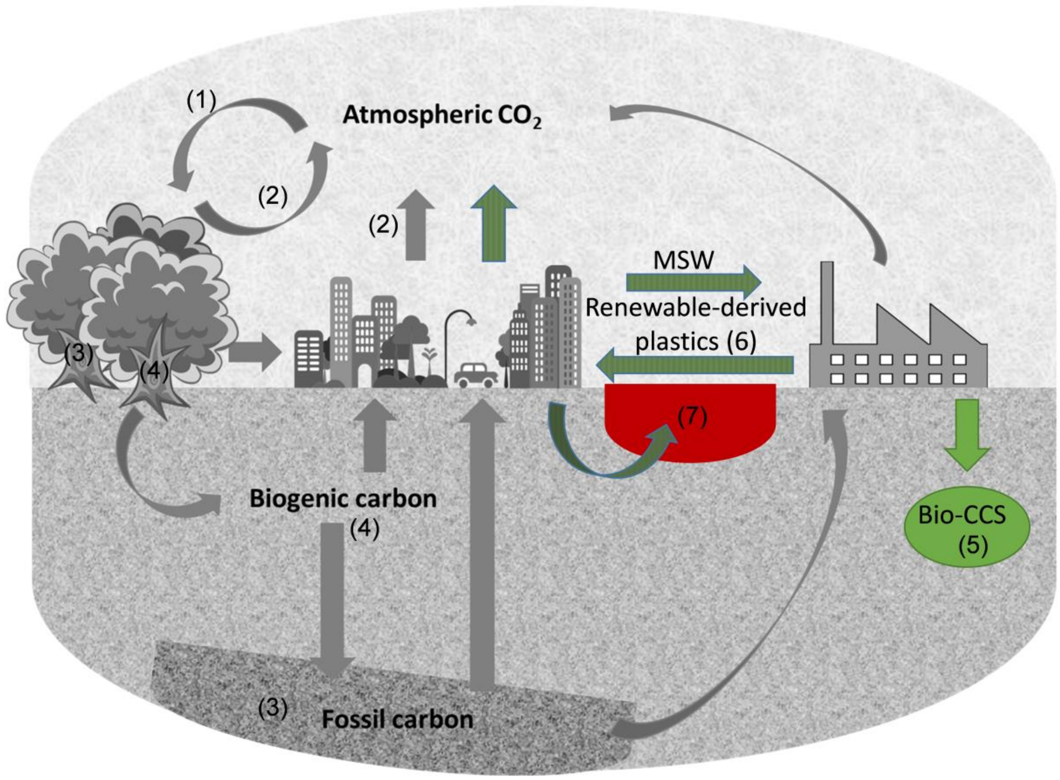


Figure 4. Carbon cycle incorporating Bio-CCS and renewable-derived plastics. Numbers refer to the different processes involved: 1. Photosynthesis, 2. Breathing and combustion, 3. Permanent storage in fossil reservoir and long-lived trees, 4. Temporal storage in living beings and soil, 5. Permanent storage in geological reservoir, 6. Temporal storage in renewable-derived plastics in use and 7. Permanent storage in renewable-derived plastics landfilled.

Table 3 shows a comparison between both GGR technologies. In the case of Bio-CCS, the stability of the process is uncertaint since technical (e.g. the need of a deeper characterization of CO_2 storage sites) and financial limitations are seriously hindering their deployment and implementation (Kemper et al., 2015, ETP, 2015, Lomax et al., 2015, Sigurjonsson et al., 2015). In line with this, an important concern with Bio-CCS is its dependence on large-scale bioenergy use, which can have adverse impacts on land use and biodiversity (IPCC, 2012). In the same way, the stability of the biogenic carbon storage in renewable-derived plastics is assessed in this thesis since it is strongly dependent of the plastic waste management scheme. However, renewable-derived plastics can be directly introduced into existing and established value chains, infrastructure and markets (Arvidsson et al., 2016, Haro et al., 2014) reducing the uncertainties about the stability of these plastics as GGR technology.

Table 3. Comparison between Bio-CCS and renewable-derived plastics.

	Regulated	**GGR**	**Stability**	**Large-scale**	**Temporality**	**Geological storage**
Bio-CCS	x	x	?	x	-	x
Renewable-derived plastics	-	x[a]	?	-[b]	x	-

[a] Proposed in this thesis.
[b] Independent of the scale.

2. Goal and scope

The aim of this thesis is the assessment of the climate benefit in the production of products (biofuels, drop-in chemicals and renewable-derived plastics) and/or services (electricity and heat) using MSW refuse as feedstock in advanced WtE plants[5]. The climate impact of existing (combustion for heat and power) and emerging (gasification for products and services) concepts of advanced WtE plants has been assessed.

The specific aims in this thesis are presented in Figure 5:

- Assessment of the GHG emissions associated to different configurations of advanced WtE plants producing biofuels and drop-in chemicals from MSW refuse (static assessment) with the possibility of Bio-CCS incorporation (Paper I).
- Assessment of the dynamic GHG emissions in an advanced WtE plant (thermochemical biorefinery) producing biofuels and electricity from MSW refuse (Paper II).
- Analysis of the climate benefit in the production of biofuels from MSW refuse in Europe considering the climate burdens from different MSW management schemes in Europe (Paper III).
- Contrasting the potential climate benefit of renewable-derived plastics from MSW refuse and Bio-CCS in advanced WtE plants (Paper IV).
- Assessment of alternative for the management of MSW refuse in incineration and gasification-based WtE plants (environmental and techno-economic analysis) (Paper V).
- Assessment of alternative for the management of MSW refuse in incineration and gasification-based WtE plants (dynamic environmental assessment) (Paper VI).

[5] In advanced WtE plants the metal recovery from bottom and/or fly ashes is possible. Metal recovery is interesting from the GHG emission assessment point of view since it involves avoided emissions from metal industry. However, metal recovery is not considered in this thesis since there are many uncertainties about the recovery potential, the type of ashes to be used and the profitability of the process. Moreover, the construction and dismantling of the WtE plants are not considered in this thesis since their impact over time is considered negligible.

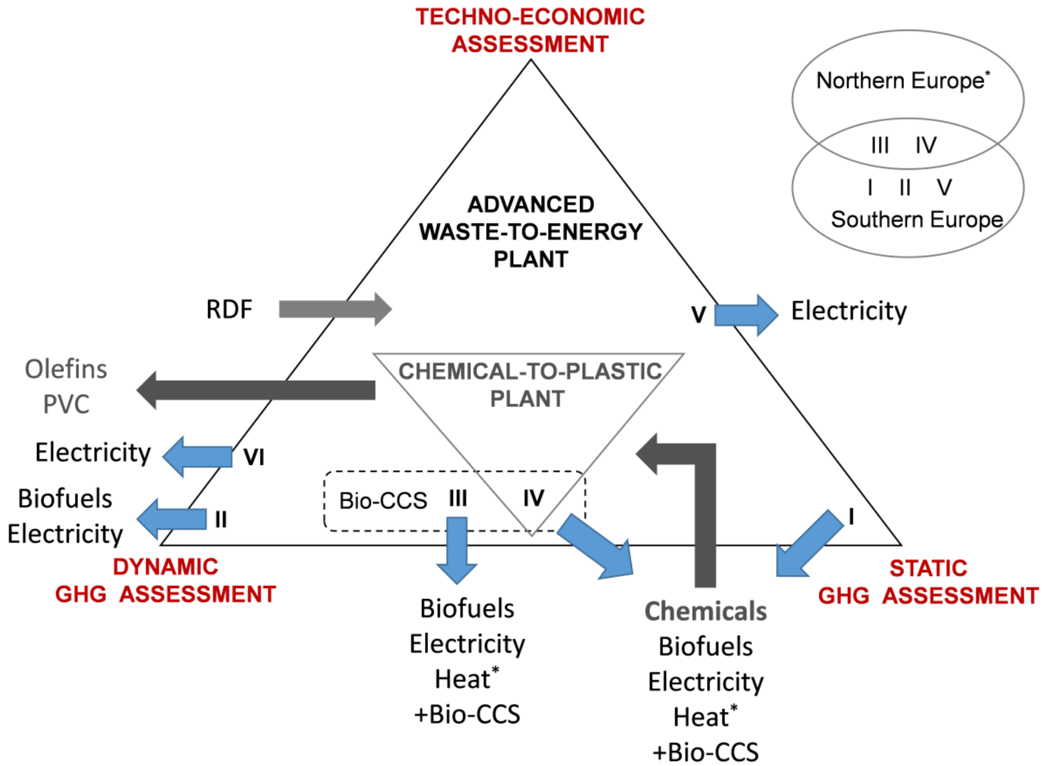


Figure 5. Overview of the appended papers and their scope. In red, the pillars of the methodology applied in this thesis. Blue flags relate each Paper to the mix of products and services assessed and green flags shows the route from chemical to plastics analyzed in Paper IV. Circles represent the geographical scope of the thesis. The asterisk means the production of district heating is only considered in Northern European countries.

2.1. Geographical scope of the thesis

The geographical scope of this thesis is Europe. In Europe, the different municipal waste management schemes can be described into two extreme cases: dominant landfilling or dominant incineration. Dominant landfilling is representative of the Southern and Eastern Europe whereas the dominant incineration is representative of the Central and Northern Europe. Spain and Sweden have been selected to represent each case (Figure 6). The availability of data and the knowledge of the situation in the countries were taken into consideration in the decision-making. Sweden was chosen since the incineration is the main way to manage the MSW refuse but also this country has a low-carbon electricity (based on nuclear energy and renewables) and heat mix (based on industrial excess heating, WtE plants and renewables). Spain was chosen since landfilling is dominant in this country and it has a high-carbon electricity mix. A great difference between Spain and Sweden is their need of district heating, in Sweden is essential during most of year whereas in Spain only some regions in the Northern need it during the winter. Because of that, the advanced WtE plant in Sweden produces district heating from excess heat in the process (Paper III and IV). In the rest of the papers, the municipal waste management scheme in Spain and particularly in the region of Andalusia was analyzed. The Spanish waste management scheme is based on recycling and landfilling and, to a lesser extent, on incineration. However, Andalusia is a region in the south of Spain where landfilling is dominant and incineration is not implemented, very similar to Southern and Eastern European countries. Hence, the implementation of WtE plants replacing landfills was analyzed for the case of Andalusia (Paper I, II and V).

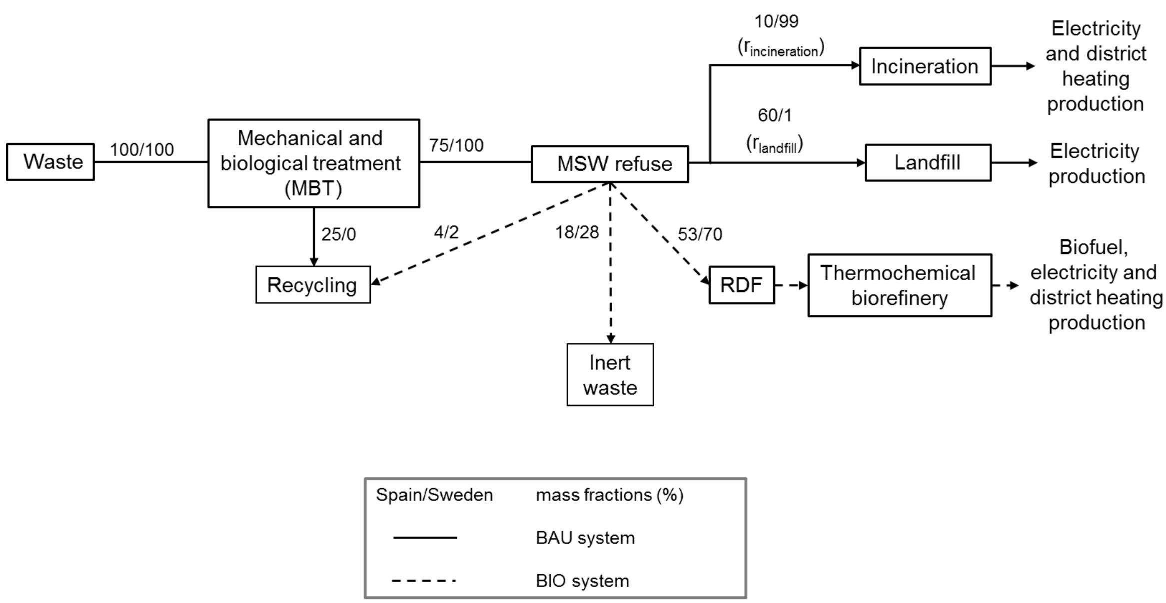


Figure 6. Waste management scheme in Spain and Sweden. In Spain, MSW refuse is the unsorted fraction coming from MBT plants whereas in Sweden, this fraction is separated at source.

3. Context of the thesis

The research conducted in this thesis started in the context of the BIOTER (*BIOrefinería TERmoquímica basada en dimethyl-ether (DME)*) project granted to BEGUS[6] for the assessment of new concepts of thermochemical biorefinery using DME as a platform chemical in 2012. One of the pillars of this project was the assessment of the GHG emissions associated to the life cycle of these thermochemical biorefineries concepts. A tool for the calculation of GHG emissions was developed and applied to these configurations using lignocellulosic biomass as feedstock. The tool was based on a simplified carbon footprint assessment called GHG balance. In 2014, a summary of the results of this tool was exhibited in the 22nd European Biomass Conference and Exhibition. From the experience in this conference and once the BIOTER project was completed emerged the interest of developing a tool for the calculation of time-integrated GHG emissions and the search and analysis of new feedstocks.

In 2015 and 2016, results of time-integrated GHG emissions using MSW refuse as feedstock in thermochemical biorefineries producing products (biofuels and/or renewable-derived drop-in chemicals), and/or services (electricity and/or heat) were presented in the 23rd European Biomass Conference and Exhibition in Vienna and in the 6th International Conference on Engineering for Waste and Biomass Valorization in Albi (France).

In parallel, an exhaustive assessment of the emissions associated to landfilling and incineration with energy recovery was carried out. The aim of this assessment was to develop a short-time alternative scenario to the thermochemical biorefinery. Therefore, a local study where incineration with energy recovery was the alternative to landfilling was economic and environmental assessed and exhibited in the 6th International Conference on Engineering for Waste and Biomass Valorization in Albi (France).

[6] Bioenergy Group of the Chemical and Environmental Engineering Department of the University of Seville headed by Prof. D. Pedro Ollero, http://grupo.us.es/bioenergia/es/.

4. Methodology

Table 4 shows a summary of the main parameters and indicators used in the methodology according to the type of GHG emission assessment (static or dynamic) in order to specify if they has been developed in this thesis or, on the contrary, they are based on literature.

Table 4. Summary of the main parameters and indicators used in this thesis.

	Parameters and/or indicators	Reference
Static GHG emission assessment	GHG balance and saving	International organizations, literature and EU regulation
	Differential GHG impact	
	Average biogenic carbon storage in renewable-derived plastics ($\bar{e}_{pool}$)[7]	Developed in this thesis
Dynamic GHG emission assessment	Time-integrated biogenic carbon storage in renewable-derived plastics over time (e_{pool})[7]	Developed in this thesis
	AGWP and AGTP	International organizations, literature and EU regulation
	DCI and CMI	Developed in this thesis

4.1. Definition of the Bioenergy (BIO) and Business As Usual (BAU) systems

In this thesis, a scheme is a set of related systems associated to a particular country, region or area land, e.g. waste management system in Sweden; whereas a system is understood as a set of related units contributing to a particular objective, e.g. reference system. In this case, every unit is an emission source, biogenic or anthropogenic, using a

[7] In the modeling of the biogenic carbon storage in renewable-derived plastics a parameter to be included in the GHG balance (static) ($\bar{e}_{pool}$) has been calculated as well as the evolution of this parameter over time (dynamic) (e_{pool}).

particular feedstock for the generation of particular product o products, e.g. an incineration plant is a unit in the reference system using MSW refuse in the production of electricity (and heat) and releasing GHG emissions to the atmosphere during the process (Figure 7). If these units belong to a conventional system in a particular region, this system is called in this thesis Business As Usual (BAU) system. The BAU system can be different between regions since the waste management is country-specific, e.g. a landfill site is a unit in the BAU system of Spain but not in Germany where MSW landfilling is negligible (EUROSTAT, 2014). On the contrary, if a system is proposed in a particular region in order to mitigate GHG emissions from the waste management, the system is called Bioenergy (BIO) system. In this case, the main unit of the BIO system is an advanced WtE plant.

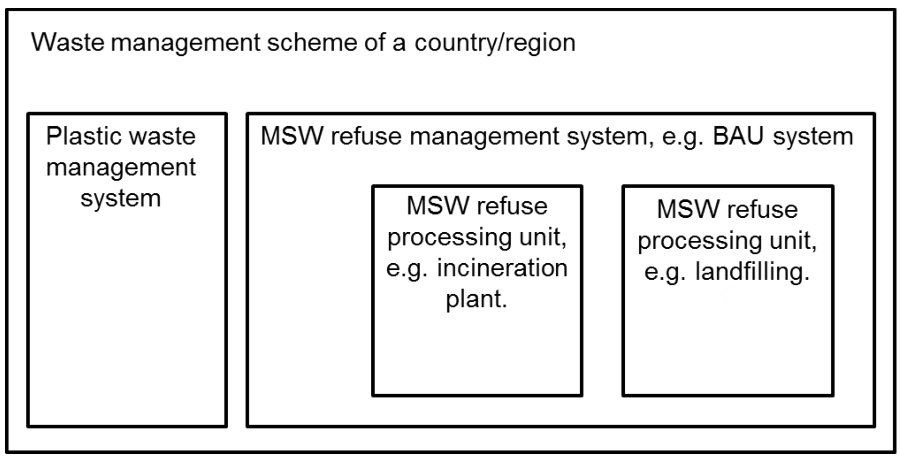


Figure 7. Relations between scheme, system and unit used in this thesis.

In a LCA study, the functional unit is a measure of the function of the studied system and it provides a reference to which the inputs and outputs of the system/process/product analyzed can be related. Typically, the functional unit can be defined for the input or the output to the e.g. the system analyzed. However, when two systems are compared in terms of an LCA study, it is crucial that the functional unit is defined for the input and the output in both systems. When the functional unit is defined for both input and output, the deficit of one or several products must be compensated from a conventional unit, e.g. electricity grid mix. Because of that, in the BIO system there may be fossil-derived emission sources. Attributional modelling is life cycle inventory (LCI) modelling frame that inventories the inputs and output flows of all processes of a system as they occur.

Modelling process along an existing supply-chain is of this type (EUR 24708 EN, 2010). In this thesis, long-term marginal processes with system expansion and substitution is applied making somehow hard to classify the study as either attributional[8] or consequential[9]. As indicated in the ILCD Handbook (EUR 24708 EN, 2010), the kind of LCA study done here could be classified as interactional. The apparent mixing of strategies in the methodology is only that apparent since a systematic approach has been followed according to current recommendation from the Joint Research Centre (JRC) as it will be shown latter. Considering the different categories usually analyzed, in this thesis only the climate change is considered. The horizon for this assessment is 100 years as the Intergovernmental Panel on Climate Change (IPCC) recommends (IPCC, 2014).

Figure 8 shows an example of the boundaries of the BIO and BAU systems where the functional unit is defined for both the input and the output of the two systems analyzed. In the BAU system, the MSW refuse is landfilled or incinerated to produce electricity and/or heat whereas fuels and/or drop-in chemicals are produced from fossil fuels. In the BIO system, MSW refuse is used in the advanced WtE plant to produce products (biofuels and/or renewable-derived plastics) and services (electricity and/or heat); and an input from electricity grid and heat mix are considered to balance the heat and/or electricity production in the BAU system. Therefore, both systems (BIO and BAU) have the same input (same amount and type of feedstock) and output (i.e. same amount and type of final products and services). The functional unit is in most cases defined using a basis of 1 MJ of output product and services from the advanced WtE plant. Therefore, substitution and system expansion are defined accordingly. Only in the Paper V the functional unit is defined using a basis of 1 ton of MSW refuse input to the system. Although in Spain the district heating might be interesting in some regions, the impact of a future use of MSW refuse in a hypothetical Spanish district heating is considered negligible. Hence, district heating is only analyzed in the Swedish case.

[8] The attributional life cycle model depicts its actual or forecasted specific or average supply-chain plus its use and end-of-life value chain. The existing or forecasted system is embedded into a static technosphere (EUR 24708 EN, 2010).

[9] The consequential life cycle model depicts the generic supply-chain as it is theoretically expected in consequence of the analyzed decision. The system interacts with the markets and those changes are depicted that an additional demand for the analyzed system is expected to have in a dynamic technosphere that is reacting to this additional demand (EUR 24708 EN, 2010).

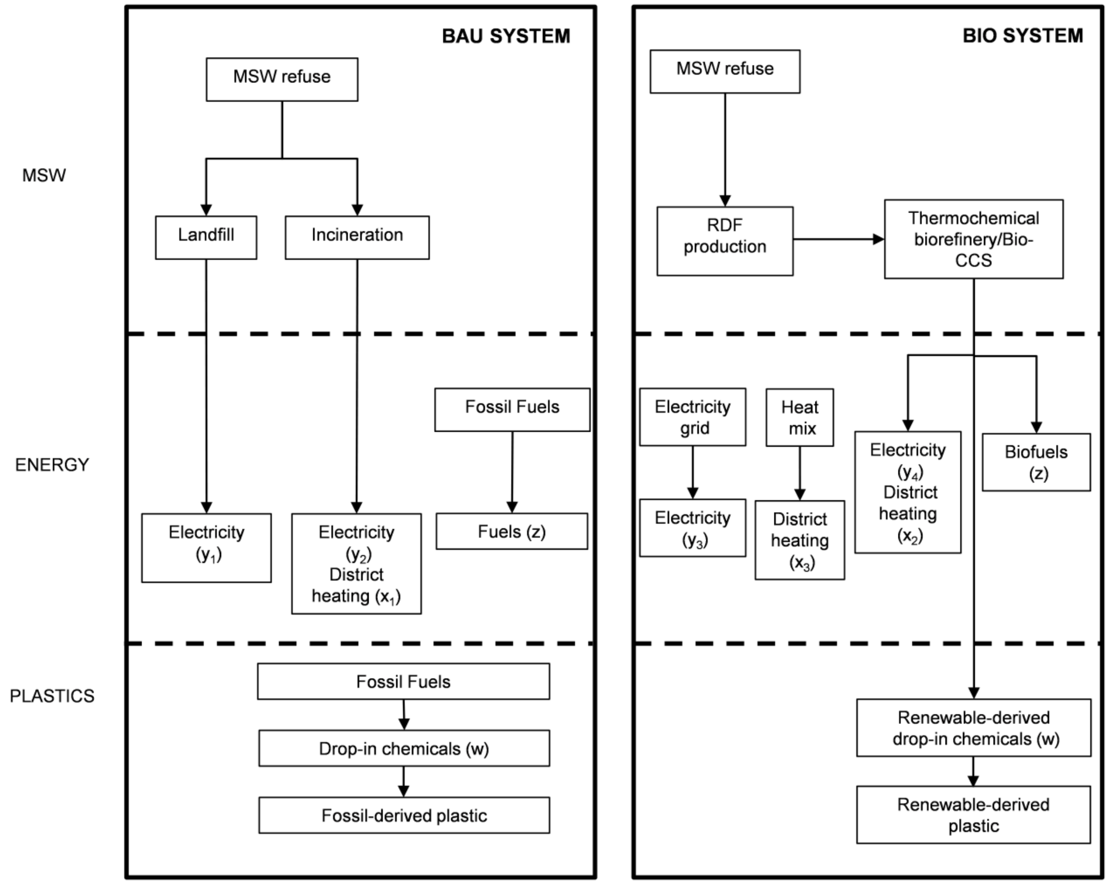


Figure 8. BIO and BAU system considered in the Paper III (MSW and energy) and Paper IV (MSW, energy and plastics). The functional unit is based on 1 MJ of products and services from the advanced WtE plant (output). In both BAU and BIO systems same amount of MSW refuse is processed and the same amount of products and services are generated.

4.2. Static GHG emission assessment

4.2.1. GHG balance and saving in the advanced WtE plant (unit)

The methodology for the calculation of the GHG balance and saving has been previously discussed using lignocellulosic biomass (i.e., thermochemical biorefinery) and the standard LCA characterization method, i.e., GWP (100), and characterization factors defined by IPCC AR5 (Haro et al., 2015). The previous methodology was adapted for the production of biofuels and services with the option of Bio-CCS incorporation to the thermochemical biorefinery. However, when using MSW refuse, it needs to be extended to include the fossil fraction in the feedstock and the production of renewable-derived plastics along with the temporal storage of biogenic carbon in renewable-derived plastics (as a negative contribution to GHG emissions). Figure 9 shows the carbon flows and pools considered in the calculation of the GHG balance. The carbon flows are emissions to the atmosphere and the carbon pools are the two alternatives of carbon storage considered in this thesis (renewable-derived plastics and Bio-CCS). The biogenic emissions are CO_2 emissions from the biogenic fraction of the MSW refuse and anthropogenic emissions are fossil emissions and non-CO_2 GHG emissions from the biogenic fraction.

Equation (1) shows how to calculate the global GHG balance for the advanced WtE plant (E_{WtE}). In the thesis, two drop-in chemicals have been modeled for the production of renewable-derived plastics: dimethyl-ether (DME), which is used to produce both biofuels ($f_{biofuel}$) and renewable-derived plastics (f_{plast}), and methyl acetate (MA), which is fully used for the production of renewable-derived plastics.

$$E_{WtE}=e_{pt}+e_p+e_t+e_{ash}+\left(x_{DME}\cdot f_{biofuel}\,(e_d+e_u)+x_{DME}\cdot f_{plast}\left(e_d+e_u+\overline{e}_{pool}\right)\right)+x_{MA}(e_d+e_u)+x_{elect}(e_d+e_u)+x_{heat}(e_d+e_u) \quad (1)$$

where f_i is the fraction of product i produced from DME (in %); x_i is the fraction of product i in the system (%); and α is the biogenic fraction in the feedstock.

RED Directive regulates the certification of energy carriers (biofuels and bioliquids) imposing a minimum GHG saving. According to RED, the saving is calculated regarding the final use of the energy carrier as a transportation fuel (biofuel), or for the generation of electricity or heat (bioliquids) (Haro et al., 2015). The GHG saving is calculated for the production of biofuel (compared to emissions from transportation fuels) using the guidelines from RED. Equation (2) is used in the calculation of the GHG saving from the production of biofuels as indicated in the European regulation (Directive 2009/28/EC).

$$\text{Saving (\%)}=\frac{EF_{biofuel}-E_{WtE}}{EF_{biofuel}} \quad (2)$$

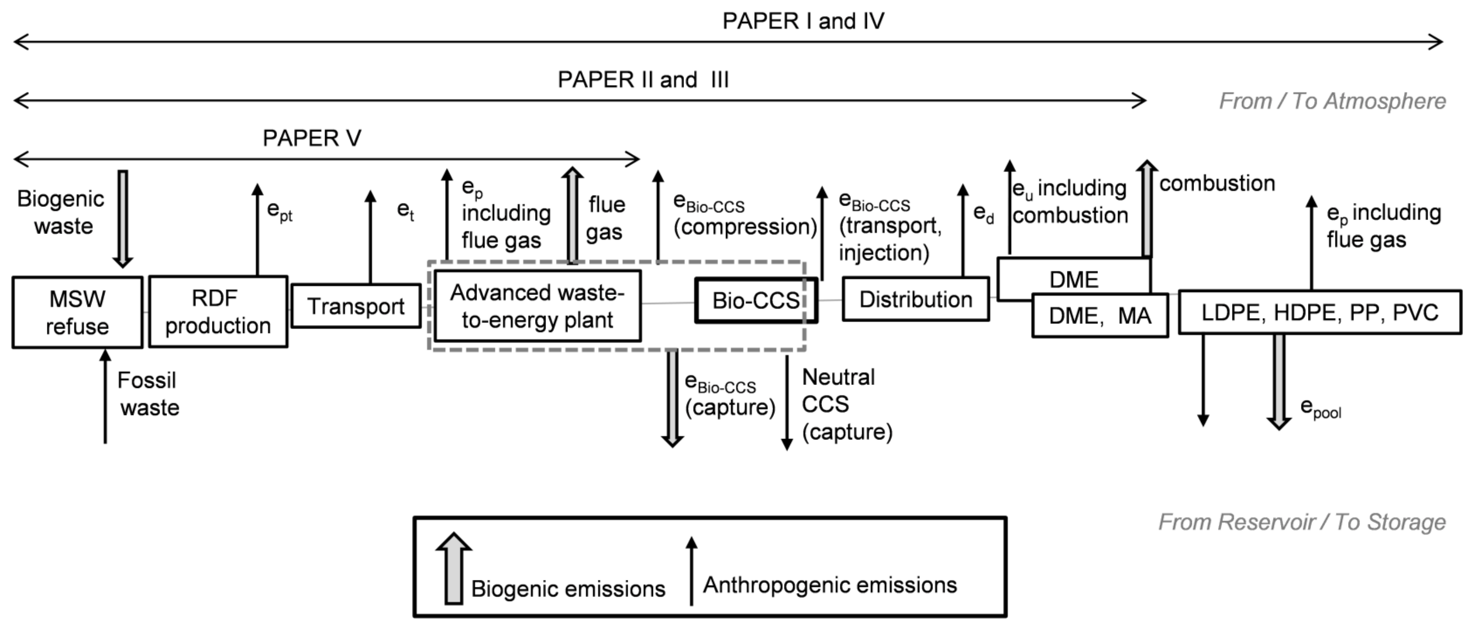


Figure 9. GHG balance associated to the life cycle of the advanced WtE plant using MSW refuse as feedstock. The modifications to the previous methodology in Haro et al. 2015 are the incorporation of the fossil fraction in the feedstock (MSW refuse) and production of renewable-derived plastics.

4.2.2. Differential GHG impact comparing BIO and BAU systems

The differential GHG impact (Equation 3) compares, in terms of GHG emissions reduction, the use of MSW refuse in advanced WtE plants producing products and services (BIO system) with the BAU system (Gaudreault, et al. 2015). It differs from the GHG balance in the consideration of the displaced and/or avoided emissions due to both material and energy substitution (i.e., it includes the burdens by means of system expansion and substitution for the BIO and BAU systems). The GHG impact of the BIO (E_{BIO}) and BAU (E_{BAU}) systems are calculated using Equation (4) and (5), respectively. Avoided emissions are represented by a fossil reference value (EF) according to the mix of products from the advanced WtE plant (Table 5). For heat, a conservative value is taken for the Swedish heat mix based on the production of district heating using waste and biomass CHP plants and from industrial excess heating (Systems Perspectives on Biorefineries, 2014).

Table 5. Fossil references for products of the advanced WtE plant.

	Spain	Sweden
$\mathbf{EF_{fuel}}$ (Directive 2015/1513/CE) (g CO_2 eq.·MJ^{-1})	90.3	
$\mathbf{EF_{olefins}}$ (Eco-profiles-PP, 2014) (g CO_2 eq.·MJ^{-1})	38	
$\mathbf{EF_{PVC}}$ (Eco-profiles, PVC, 2005) (g CO_2 eq.·MJ^{-1})	94	
$\mathbf{EF_{electricity}}$ (EUR 27215 EN) (g CO_2 eq.·MJ^{-1})[1]	110.7	17.5
$\mathbf{EF_{heat}}$ (Systems Perspectives on Biorefineries, 2014) (g CO_2 eq.·MJ^{-1})	-	-68

[1]In Sweden the average CO_2 emissions per MWh of electricity is 0.063 t fossil CO_2/MWhe) (EUR 27215 EN) and 0.398 t fossil CO_2/MWhe in Spain (EUR 27215 EN).

$$\text{Differential GHG impact}=E_{BIO}-E_{BAU} \tag{3}$$

$$E_{BIO}=E_{WtE}\cdot(y_4+z_2+w_2+x_2)+EF_{electricity}\cdot y_3+EF_{heat}\cdot x_3 \tag{4}$$

$$E_{BAU}=EF_{fuel}\cdot z_1+ EF_{plast}\cdot w_1+(\textstyle\sum_i E_i\cdot r_i)\cdot(y_1+y_2+x_1) \tag{5}$$

where i is landfill and/or incineration.

$E_{landfill}$ represents the GHG emissions from the landfill, which depends on the behavior of landfilled materials. Although IPCC and EPA establish default values, some parameters are country-specific and even vary between regions. $E_{incineration}$ represents the GHG emissions from MSW refuse incineration. The results in terms of grams of CO_2 equivalent per ton of MSW refuse are corrected by the unit efficiency (Equation 6) to use the same functional unit in both systems: 1 MJ of total products and services from the advanced WtE plant. The purpose of this correction factor is to include the impact of the different conversion efficiencies in the BAU system compared with the BIO system.

$$E_i = \frac{g\ CO_2\ eq}{t\ MSW\ refuse} \cdot \frac{t\ MSW\ refuse/year}{(MJoutput/year)^{*}\left(\frac{\eta_i}{\eta\ biorefinery}\right)} \quad (6)$$

where i is landfill or incineration

Figure 10 shows the how the main parameters used in the static assessment are interconnected.

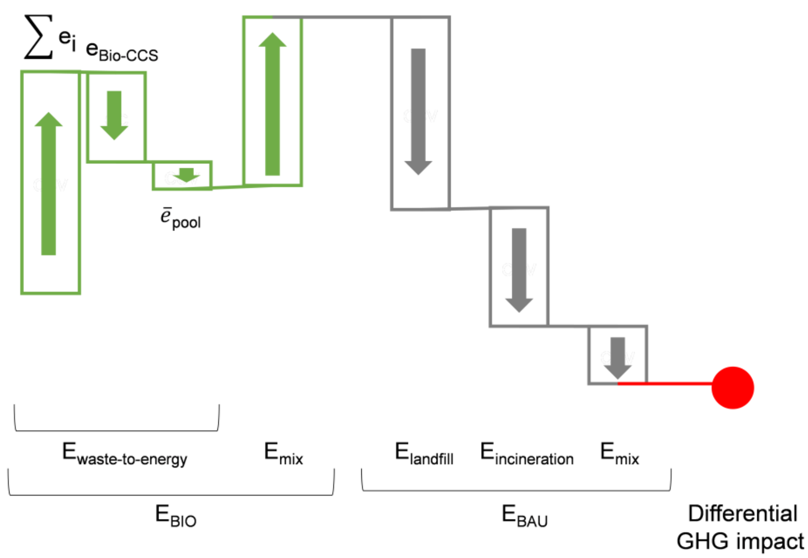


Figure 10. Overview of the differential GHG impact for the BIO and BAU systems in terms of the GHG balances for each unit.

4.3. Dynamic GHG emission assessment

4.3.1. AGWP and AGTP in the BIO and BAU systems

This methodology has been applied to each unit of the BIO and BAU systems, i.e., landfilling, incineration plant, advanced WtE plant and fossil references (fossil fuels, fossil-derived plastics and electricity and heat mix) and further integrated in four parameters (AGWP and AGTP for both systems). The calculations are carried out per year (t) up to a time horizon of 100 years (T).

To calculate the time-dependent climate mitigation potential, each individual Well Mixed Greenhouse gas (WMGHG), i.e., CO_2, CH_4 and N_2O, has been modelled in each unit. The WMGHG have lifetimes long enough to be (relatively) homogeneously mixed in the troposphere. The emissions to the atmosphere of each individual WMGHG from the management of the MSW refuse produced in a year are called pulse emissions (E) (g CO_2 eq.·MJ^{-1}) (IPCC, 2013, Sathre and Gustavsson, 2012). The pulse emissions undergo an atmospheric decay (R) over time (calculated up to year 100) due to the natural dynamic of the atmosphere (C and N cycles) called in this thesis the GHG remaining (rGHG) (g CO_2 eq.·MJ^{-1}) (Equations 7-10, Table 6) (IPCC, 2013, Sathre and Gustavsson, 2012). The sum of the pulse emissions in year i and the GHG remaining in the atmosphere from year 1 to (i-1) is the cumulative GHG (GHG_c) (g CO_2 eq.·MJ^{-1}) (Equation 11).

$$R_{CO_2}(t) = a_0 + \sum_{i=1}^{3} a_i \cdot e^{-t/\tau_i} \tag{7}$$

$$R_{CH_4}(t) = e^{-t/\tau_{CH_4}} \tag{8}$$

$$R_{N_2O}(t) = e^{-t/\tau_{N_2O}} \tag{9}$$

Table 6. Parameters used for the calculation of R in Equations 8 to 10 (IPCC; 2013, Giuntoli et al., 2015b).

a_0	a_1	a_2	a_3	τ_1	τ_2	τ_3	τ_{CH4}	τ_{N2O}
0.2173	0.224	0.2824	0.2763	394.4	36.54	4.304	12.4	121

$$rGHG(t) = \sum_{1}^{t} E(t-1) \cdot R(t) \tag{10}$$

$$GHG_c = E(t) + rGHG(t) \tag{11}$$

The CO_2 concentration (ppmv in the atmosphere (Equation 12[10], Table 7) and cumulative CH_4 and N_2O are used to calculate the Radiative Forcing (RF) (W·m^{-2}·kg MSW $refuse^{-1}$) (Equations 13-15, Table 8) (IPCC; 2013, Giuntoli et al., 2015b). RF is a simple measure for both quantifying and ranking the many different influences on climate change (natural and anthropogenic). RF is divided by the kilograms of MSW refuse per year. The

[10] In order to calculate AGTP, the CO_2 concentration is obtained from the pulse emission and not from the cumulative CO_2, i.e., in equation 13 $(CO_2)_t$ is replaced by E_i.

cumulative Radiative Forcing or Absolute Global Warming Potential (AGWP) ($W \cdot year \cdot m^{-2} \cdot kg$ MSW refuse^{-1}) is calculated integrating the RF over a defined time horizon (IPCC; 2013, Giuntoli et al., 2015b, Pingoud et al., 2012) (Equation 16). Absolute Global surface Temperature change Potential (AGTP) ($K \cdot kg$ MSW refuse^{-1}) is the global mean surface temperature change at a chosen point in time in response to an emission pulse (Equations 17 and 18, Table 9) (Giuntoli et al., 2015b). According to IPPC, the uncertainty range for RF and AGWP is ±24% and ±26% respectively (IPCC, 2013, Joos et al., 2013). Final AGWP and AGTP are obtained for the BIO and BAU system considering the share of energy of each unit into each system.

$$[CO_2]=\frac{(CO_2)c \cdot 10E3}{\text{atmosphere mass}} \cdot \frac{\text{dry air molecular mass}}{CO_2\text{molecular mass}} \tag{12}$$

Table 7. Parameters used in the calculation of $[CO_2]$ in Equation 12.

Atmosphere mass (kg)	5.14E+18
Dry air molecular mass ($g \cdot mol^{-1}$)	28.97
CO_2 molecular mass ($g \cdot mol^{-1}$)	44.01

$$RF_{CO_2}=5.35 \cdot \ln(1+\frac{[CO_2]}{C_0}) \tag{13}$$

$$RF_{CH_4}=(1+f_1+f_2) \cdot A_{CH4} \cdot CH_{4c} \tag{14}$$

$$RF_{N_2O}=(1-0.36 \cdot (1+f_1+f_2) \cdot \frac{RE_{CH4}}{RE_{N2O}}) \cdot A_{N2O} \cdot N_2O_c \tag{15}$$

where

C_0: CO_2 concentration in the atmosphere (ppmv) (IPCC, 2013)

f_1 and f_2: correction factor due to effects on ozone and stratospheric H_2O respectively (IPCC, 2013)

A_i: radiative forcing per unit mass ($W \cdot m^{-2} \cdot kg$ MSW refuse^{-1}) (IPCC, 2013)

RE_i: radiative efficiency of gas I on volume basis ($W \cdot m^{-2} \cdot ppb^{-1}$) (IPCC, 2013)

Table 8. Parameters used for the calculation of RF in Equations 13-15 (IPCC; 2013, Giuntoli et al., 2015b).

Parameter	Value	Unit
A_{CH4}	1.28E-13	$W \cdot m^{-2} \cdot ppb^{-1}$
RE_{CH4}	3.63E-4	$W \cdot m^{-2} \cdot kg^{-1}$
A_{N2O}	3.85E-13	$W \cdot m^{-2} \cdot ppb^{-1}$
RE_{N2O}	3.00E-3	$W \cdot m^{-2} \cdot kg^{-1}$
f_1	0.5	-
f_2	0.15	-

$$AGWP(t)= \int_0^t RF\,(t')dt' \tag{16}$$

$$AGTP(t)= \int_0^t RF\left(t'\right) \cdot \, \delta T\left(t-t'\right)dt' \tag{17}$$

$$\delta T= \sum_{i=1}^{2} \frac{ci}{di} \cdot \exp\left(-\frac{t}{di}\right) \tag{18}$$

Table 9. Parameters used for the calculation of AGTP in Equation 18 (IPCC; 2013, Giuntoli et al.,2015b).

Parameter	1st term	2nd term	unit
c_i	0.631	0.429	$K \cdot W^{-1} \cdot m^{-2}$)
d_i	8.4	409.5	years

4.3.2. CMI and DCI comparing BIO and BAU systems

In this thesis, two parameters are proposed and designed in the dynamic assessment: climate mitigation index (CMI) and differential climate impact (DCI). Table 10 gives a comparison between the CMI and the DCI.

Table 10. Comparison between the parameters used in the dynamic GHG emission assessment: CMI and DCI.

	Climate Mitigation Index (CMI)	**Differential Climate Impact (DCI)**
Based on	AGWP (cumulative)	AGTP (instantaneous)
Units	--	$K \cdot kg^{-1}$ MSW refuse
Emissions included	Biogenic and anthropogenic emissions	Anthropogenic and non-CO_2 biogenic emissions
Comparison	BIO and BAU system for the same region	Different regions and GGR technologies
Result	Cumulative climate mitigation for a specific region	Climate benefit at a specific time
Used in this thesis	Paper III and VI	Paper III, IV and VI

4.3.2.1. Climate mitigation impact (CMI)

The climate mitigation index (CMI) assesses the mitigation potential of producing products and services from MSW refuse instead of following current MSW management schemes. The CMI is dimensionless. For each region assessed (Spain or Sweden), the feedstock is identical independently of how the MSW refuse is disposed of. Therefore, since the re-absorption factor is equal in both systems, all GHG emissions (CO_2 included) are counted from the MSW refuse for both biogenic and fossil origin. The CMI is calculated according to Equation 19 (Pingoud et al., 2012, IEA, 2012). The values of AGWP for BIO and BAU systems are calculated from the annual emissions of each WMGHG as explained elsewhere (Giuntoli et al., 2015a, IPCC, 2013, Sathre and Gustavsson, 2012).

Regarding CMI values, it is possible to compare the behavior of the BIO system with the current MSW management, and production of products and services (i.e., the BAU system) for a specific region (Scheme 1, Figure 11). Since the CMI is based on the AGWP values, this comparison gives the cumulative climate mitigation of producing products and services. Therefore, if the CMI reaches a positive value at a certain time, it means at this time there will be no accumulated climate benefit, i.e., the BIO system becomes worse than the BAU system. For negative CMI values, there is an accumulated climate benefit, until the CMI reaches -1 when then the BIO system has no emissions of WMGHG.

$$CMI=\frac{(AGWP_{BIO}-AGWP_{BAU})}{AGWP_{BAU}} \tag{19}$$

$$CMI>0 \rightarrow AGWP_{BIO}>AGWP_{BAU} \rightarrow \text{Climate worsening}$$

$$CMI=0 \rightarrow AGWP_{BIO}=AGWP_{BAU} \rightarrow \text{Climate neutral}$$

$$-1<CMI<0 \rightarrow AGWP_{BIO}<AGWP_{BAU} \rightarrow \text{Climate mitigation}$$

$$CMI=-1 \rightarrow AGWP_{BIO}=0 \rightarrow \text{BIO is climate neutral}$$

(Scheme 1)

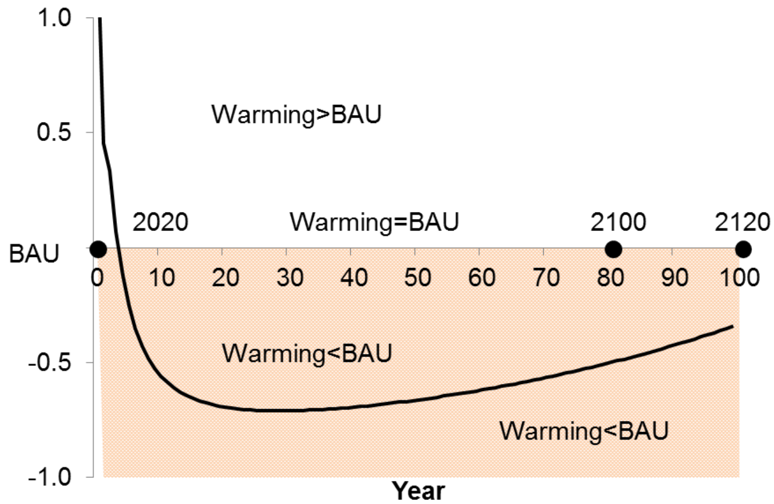


Figure 11. Graphical explanation of a time-integrated GHG emission profile using the CMI indicator.

4.3.2.2. Differential climate impact (DCI)

The differential climate impact (DCI) measures the climate benefit in the production of products and services from MSW refuse in order to compare the results of regions with different waste management schemes. The units of DCI are $K \cdot kg$ MSW refuse^{-1}. In different regions, feedstock composition differs and therefore the biogenic emissions cannot be modelled as for the CMI (Giuntoli et al., 2015a). As mentioned above, in this case it is only possible to account for fossil carbon and biogenic non-CO_2 emissions. In the same way, the biogenic carbon stored in landfill and plastics; and from Bio-CCS incorporation are modelled as negative contributions. The DCI is based on the AGTP metric as the surface temperature response to the replacement of BAU by BIO (Equation 20). The values of AGTP for BIO and BAU systems are calculated from the annual emissions of each WMGHG as explained elsewhere (Giuntoli et al., 2015a, IPCC, 2013, Sathre and Gustavsson, 2012).

Regarding the values of the DCI, a direct comparison of two different regions can be made. Since the DCI is based on AGTP values, the comparison gives the climate benefit at a specific time; an instantaneous comparison that is not biased by the accumulated effect of previous WMGHG emissions. Therefore, if the DCI of one region is lower than in another region, it means that there is a larger climate benefit in the production of biofuels and/or renewable-derived drop-in chemicals in this region at a specific time.

$$DCI = AGTP_{BIO} - AGTP_{BAU} \qquad (20)$$

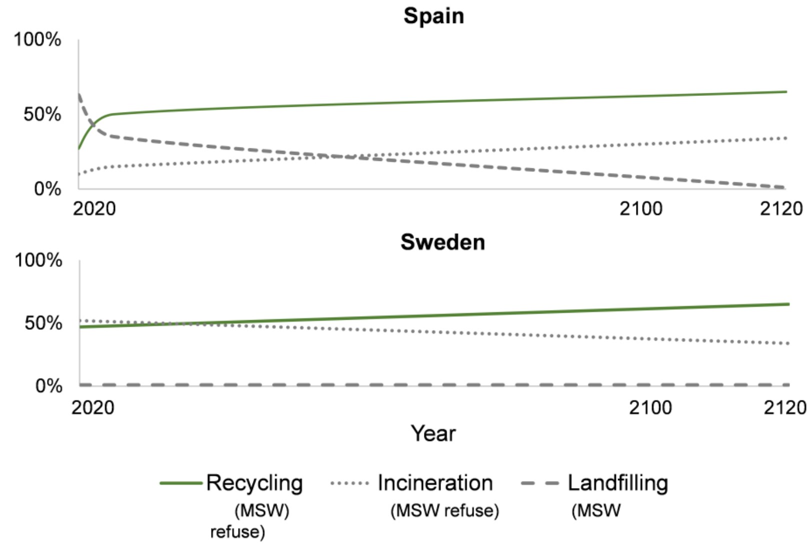
4.4. Scenarios

Two scenarios for the evolution of the BAU system (MSW refuse management, product and services production) are considered.

- Scenario 1. The production in the advanced WtE plant (unit) is continuous. In this scenario, we assume that the MSW management scheme and products (including plastics) and services production do not change for the whole period (for both BIO and BAU systems). This scenario applies for both the static and dynamic assessments. The aim of this scenario is the comparison between the static and the dynamic assessment since the static assessment cannot include any evolution of the MSW refuse management or the products and services production.
- Scenario 2. It considers an evolution of the MSW refuse management to the legal targets and recommendations set by the European Commission. This scenario applies only for the dynamic. This evolution brings a landfill-banned BAU system and would be closer to the future evolution of MSW management in Europe for both MSW refuse and plastic waste (Figure 12). Considering the selected regions, the targets set in the landfill and the waste framework Directives have been already achieved in Sweden but not in Spain. Therefore, Spain should reduce the amount

of biodegradable municipal waste in MSW refuse and its landfilling rate (Directive 2008/98/EC). This scenario also considers an evolution in the energy mix for both Spain and Sweden according to forecasts of the European Commission (Figure 13).

(1)



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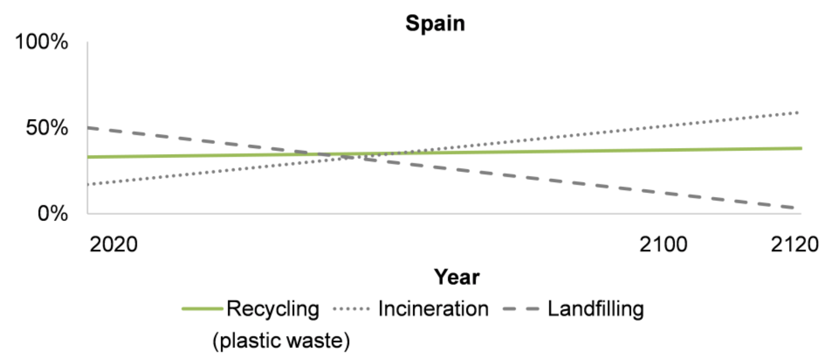


Figure 12. Targets to be achieve in Scenario 2 in the MSW refuse management (1) for Spain and Sweden (Aracil et al., 2017); and in the plastic waste management (2) for Spain.

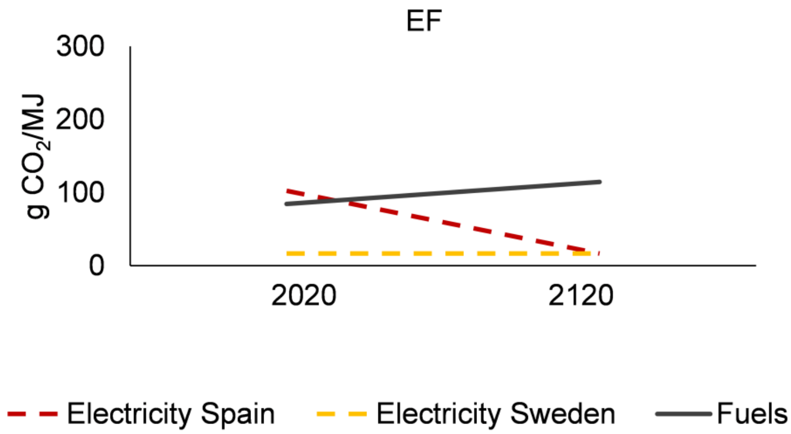


Figure 13. Evolution of emissions from the energy mix (electricity and fuels) in scenario 2 for both Spain and Sweden. For clarity, the yearly emissions of each WMGHG are combined and expressed in g CO_2 eq./MJ (Aracil et al., 2017).

4.5. Modeling of biogenic carbon storage in renewable-derived plastics

The modeling of the storage of biogenic carbon in renewable-derived plastics is carried out defining three parameters. The first represents the biogenic carbon captured in products still in use and recycled products, i.e. plastic materials in the market ($e_{material}$), the second represents the biogenic carbon captured in landfilled products ($e_{landfill}$). Both $e_{material}$ and $e_{landfill}$ are an additional carbon pool in the calculations. The last represents the anthropogenic emissions to the atmosphere (carbon flow) from the combustion of the plastic waste (e_{energy}). In Figure 14 the evolution along the time of each parameter is shown, the impact of the $e_{material}$ in the carbon pool is higher than this 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.

For the modelling of biogenic carbon in renewable-derived plastics, two streams must be known, products still in use in the market (P) and plastics becoming waste (W). Short-life plastic products ($P_{short\text{-}life\ plastics}$) are considered to become waste in less than a year, however, the most of long-life plastics usually remain in the market for more than one year. Therefore, Equation (21) gives the fraction of the long-life plastic products becoming waste

($W_{long\text{-}life\ plastics}$) in one year. Long-life plastic products still in use ($P_{long\text{-}life\ plastics}$) are 1-$W_{long\text{-}life\ plastics}$.

$$W_{long-life\ plastics} = \frac{W - P_{short-life\ plastics}}{P_{long-life\ plastics}} \tag{21}$$

In the year zero, $e_{material}$ represents the input of new plastic products in the market in terms of g CO_2 eq.·MJ^{-1} of plastics (C input) whereas $e_{landfill}$ and e_{energy} have a zero value. From year 1 to 100, $e_{landfill}$ and e_{energy} increase since more and more plastic waste is landfilled or energy recovered and, therefore, $e_{material}$ decreases. Equations (22) to (25) show the calculations:

- Year 0:

$$e_{material} = C\ input \cdot \alpha \tag{22}$$

where α is the biogenic fraction of the MSW refuse (%)

- Successive years for short-life plastic materials:

$$e_{material\ (year\ i)} = e_{material\ (year\ i-1)} \cdot W_{short-life\ plastics} \cdot r_{recycling} \tag{23}$$

The parameter $e_{landfill\ (year\ i)}$ is calculated as $e_{material\ (year\ i)}$ replacing $r_{recycling}$ by $r_{landfilling}$.

$$e_{energy\ (year\ i)} = e_{material\ (year\ i-1)} \cdot \frac{\beta}{\alpha} \cdot W_{short-life\ plastics} \cdot r_{energy\ recovery} \tag{24}$$

- Successive years for long-life plastic materials:

$$e_{material\ (year\ i)} = e_{material\ (year\ i-1)} \cdot (1 + W_{long-life\ plastics} \cdot (r_{recycling} - 1)) \tag{25}$$

The other two equations ($e_{landfill}$ and e_{energy}) differ from those of short-life plastic materials changing $W_{short\text{-}life\ plastics}$ by $W_{long\text{-}life\ plastics}$.

Finally, the parameter e_{pool} is calculated (Equation 26):

$$e_{pool\ (year\ i)} = (e_{material} + e_{landfill} + e_{energy})_{year\ i} \tag{26}$$

To introduce these data in a GHG balance is necessary to obtain a single value; therefore, all the values are added in the parameter $\bar{e}_{pool}$ defined as the average biogenic carbon storage in plastics for 100 years (Equation 27):

$$\overline{e_{pool}} = \bar{x}\,(e_{pool\,(from\,year\,0\,to\,100)}) \tag{27}$$

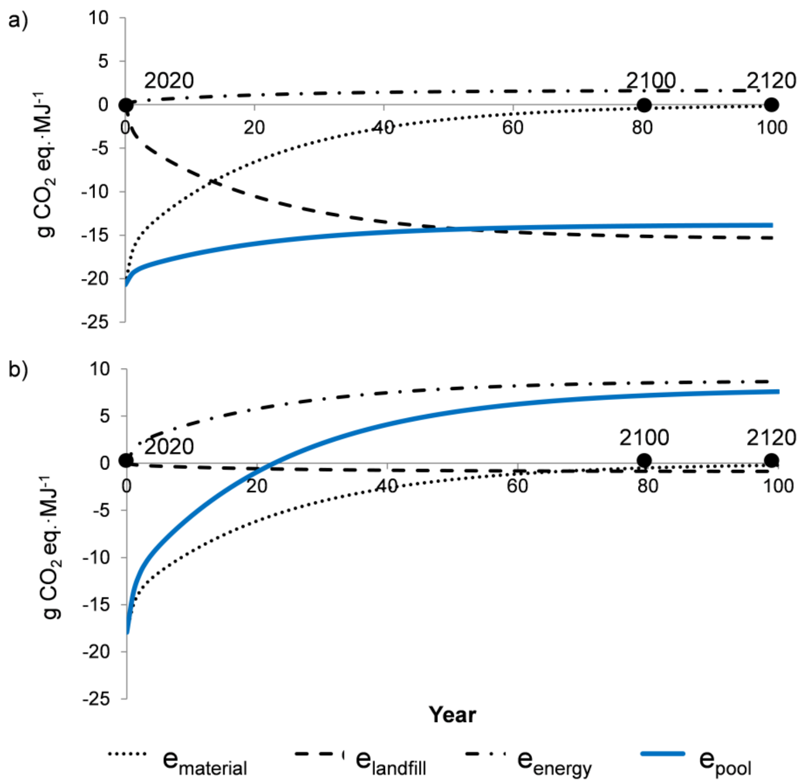


Figure 14. Evolution of the carbon flow (e_{energy}) and carbon pools ($e_{landfill}$ and $e_{material}$) for 100 years in terms of g CO_2 eq.·MJ^{-1} of plastic in Spain (a) and Sweden (b) from a carbon input in the year 0. Blue line represents the net cumulative biogenic carbon pool (e_{pool}).

4.6. Techno-economic assessment

A techno-economic assessment is carried out only for some cases, i.e., comparison of conventional and advanced WtE plants using MSW refuse in Andalusia. The advanced WtE plant is limited to the production of electricity for the techno-economic assessment.

The most extended technologies for both conventional and advanced WtE pants have been selected, moving grate and fluidized bed reactors.

The techno-economic assessment is calculated using common guidelines in process engineering. Total Investment Costs (TIC) and Total Operational Costs (TOC) are calculated using the parameters in Table 11. Equipment costs are taken from literature but updated using CEPCI parameter. In the case of the advanced WtE plant, TOC is calculated as the sum of fixed and variable costs. Fixed costs involve maintenance, insurance, labor and management, and services. These costs are calculated as a fraction of TIC. Variable costs entail RDF production, residues disposal and commodities consumption.

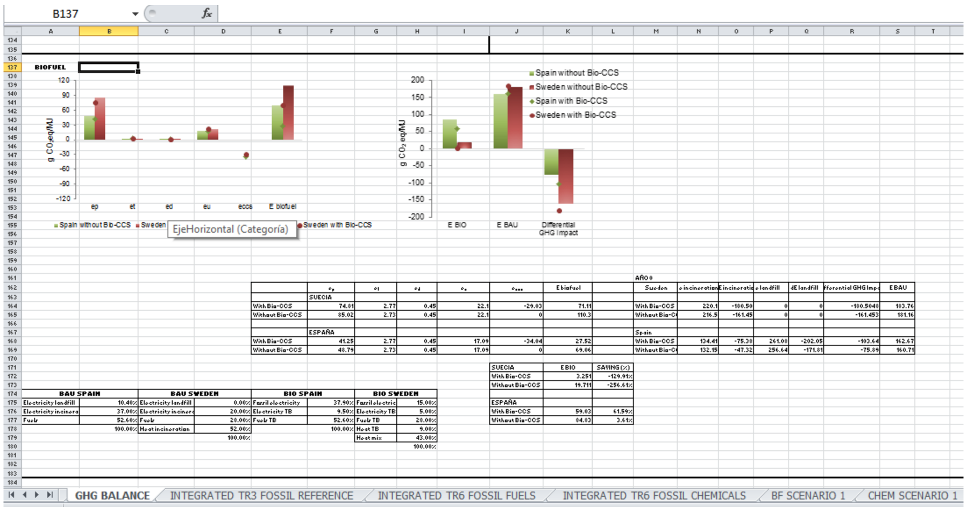
Table 11. Parameters used in the techno-economic assessment.

Inflation	2%
Capital cost	5.5%
Hours/year	7500
Corporation tax	30% of taxable (incineration) 25% of taxable (gasification)
Gate fee	Variable
Scale factor	0.6
Lang factor	3.1
Amortization years	15 years (conventional WtE) 10 years (advanced WtE)

4.7. Software used in the calculations of static and dynamic GHG emission assessment

In this thesis, the calculations and graphs have been run and plotted using a series of spreadsheets as shown in Figure 15. Thirty-five spreadsheets were developed in order to complete all the calculations required for the achievement of the results.

(1)



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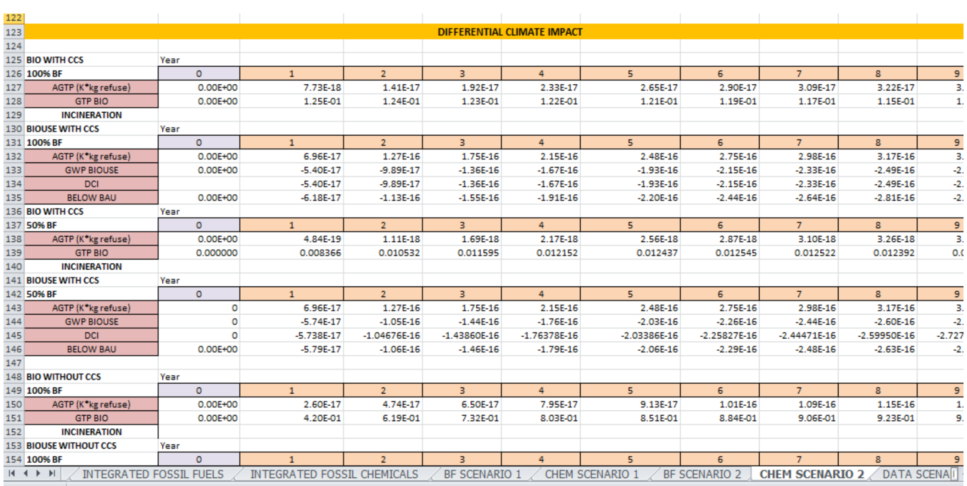


Figure 15. Overview of the spreadsheets made in this thesis for the next calcualtions: GHG balance and diffeential GHG impact (a), AGWP and AGTP (b); differential climate impact (c) and modeling of renewable-derived plastics (d).

5. Summary of the case studies

Table 12. Definition of the case studies in papers appended.

PAPER	STUDY CASES		BIO-CCS/ RENEWABLE-DERIVED PLASTICS	PRODUCTS/SERVICES	ASSESSMENT	SCENARIO	GEOGRAPHICAL SCOPE	REFERENCE SYSTEM
I	Thermochemical biorefinery	A	-/-	Biofuels and electricity	GHG balance and GHG saving	1	Andalusia	--
		B	-/+	Biofuels, renewable-derived chemicals and electricity				
		C	+/-	Biofuels and electricity + Bio-CCS				
II	Thermochemical biorefinery		-/-	Ethanol and electricity	Time-integrated GHG emission assessment	1	Andalusia	Landfill
III	Thermochemical biorefinery	w/- Bio-CCS	+/-	Ethanol, DME, electricity and heat	GHG balance, GHG saving and time-integrated GHG emission assessment	1 and 2	Spain and Sweden	Complete (BAU system)
		w/o Bio-CCS	-/-					
IV	Thermochemical biorefinery	w/- Bio-CCS	+/+	DME, olefins, PVC, electricity and heat	GHG balance, differential GHG impact and time-integrated GHG emission assessment	2	Spain and Sweden	Complete (BAU system)
		w/o Bio-CCS	-/+					
V	Incineration-based WtE plant	GC/SRC[11]	-/-	Electricity	Techno-economic and differential GHG impact	1	Southern Europe	Landfill
	Gasification-based WtE plant	FBG/ICE						
		FBG/ORC						
		GG/SRC						
VI	Advanced WtE plant	FBG/ICE	-/-	Electricity	Time-integrated GHG emission assessment	1 and 2	Southern Europe	Landfill
		FBG/ORC						
		GG/SRC						

[11] Incineration is considered into the BAU system for Sweden in Paper III and IV but in the paper V is a study case since incineration is negligible in Southern Europe, in this case, the impact of the replacement of the landfilling for a incineration and gasification-based WtE plant is assessed.

6. Results and discussion

6.1. Static assessment

Table 13 shows the results for the GHG balances and GHG saving in the BIO and BAU systems. The emissions from the advanced WtE plant (E_{WtE}) decrease when renewable-derived plastics are produced since these plastics have not associated emissions from their use (e_u) in comparison with biofuels and they store biogenic carbon (e_{pool}). Emissions from landfilling and incineration are always higher than those from the advanced WtE plant. In the case of the landfill, the methane emissions profile makes the landfill very pollutant since methane has a high warming potential (25 times more than CO_2, IPCC, 2015). However, incineration has emissions constant over time and their replacement for an advanced WtE plant involves a high climate benefit at a large time. For the storage of the biogenic carbon in renewable-derived plastics ($\bar{e}_{pool}$), their impact in the static assessment is much reduced compared with Bio-CCS.

Table 14 shows some results of the differential GHG impact. The differential GHG impact is always negative revealing the current MSW management scheme releases more GHG emissions than any alternative BIO system analyzed in this thesis. The replacement of landfilling for WtE plants involves a clear advantage when static GHG emission assessment is carried out. However, the replacement of the BAU system for the BIO system carries a higher differential GHG impact in dominant-incineration countries, i.e. Sweden. This is mainly because of the impact of the landfill for both methane emissions and biogenic carbon storage in the landfill.

The production of renewable-derived plastics (Paper IV) in the advanced WtE plant involves a similar or higher differential GHG impact since emissions in the BIO system are lower than in the case of biofuel production (Paper III). In Paper V, the BIO system based on fluidized-bed gasifier and internal combustion engine (FBG/ICE) achieves the higher differential GHG impact. Results are better for advanced BIO system based on gasification than for conventional BIO system based on incineration except in the case of the configuration FBG/ORC whose result is very similar to incineration.

Table 13. Results of the GHG balance and saving in the case studies.

	Paper V (kg CO_2 eq./t MSW refuse)				Paper III (g CO_2 eq./MJ output)				Paper IV (g CO_2 eq./MJ output)	
	Conventional BIO system	Advanced BIO system			w/- Bio-CCS		w/o Bio-CCS		w/- Bio-CCS	
		FBG/ICE	FBG/ORC	GG/SRC	Spain	Sweden	Spain	Sweden	Spain	Sweden
e_{pt}	-		(4.9)		41	75	49	85	41	62
e_p	324	280	270	280						
e_t	4.1	-	-	-	2.8		2.7		3.3	
e_{ash}	3.2		5		-	-	-	-	-	-
e_d	-	-	-	-	0.5		0.5		0	
e_u	-	-	-	-	17	22	17	22	8	7
$e_{Bio\text{-}CCS}$	-	-	-	-	-33	-29	-	-	-36	-31
$\bar{e}_{pool}$	-	-	-	-	-	-	-	-	-9	2
E_{WtE}	-	287	277	287	28	71	69	110	4	43
$E_{landfill}$		454			261	-	257	-	338	-
$E_{incineration}$	331	-	-	-	134	220	132	217	173	219
GHG Saving (%)	-	-	-	-	61	-130	4	-257	-	-

Table 14. Results of the differential GHG impact in the case studies.

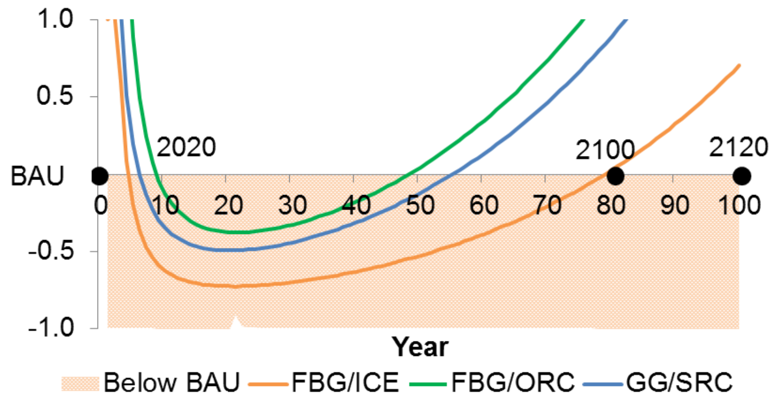
	Paper V (kg CO_2 eq./t MSW refuse)				Paper III (g CO_2 eq./MJ output)				Paper IV (g CO_2 eq./MJ output)	
	Conventional BIO system	Advanced BIO system			w/- Bio-CCS		w/o Bio-CCS		w/- Bio-CCS	
		FBG/ICE	FBG/ORC	GG/SRC	Spain	Sweden	Spain	Sweden	Spain	Sweden
E_{BIO}	331	287	277	287	59	3	85	20	33	-11
E_{BAU}	454				163	184	161	181	134	183
Differential GHG impact	-215	-340	-211	-226	-103	-181	-76	-161	-101	-194

6.2. Dynamic assessment: CMI and DCI

6.2.1. Climate mitigation index (Paper III)

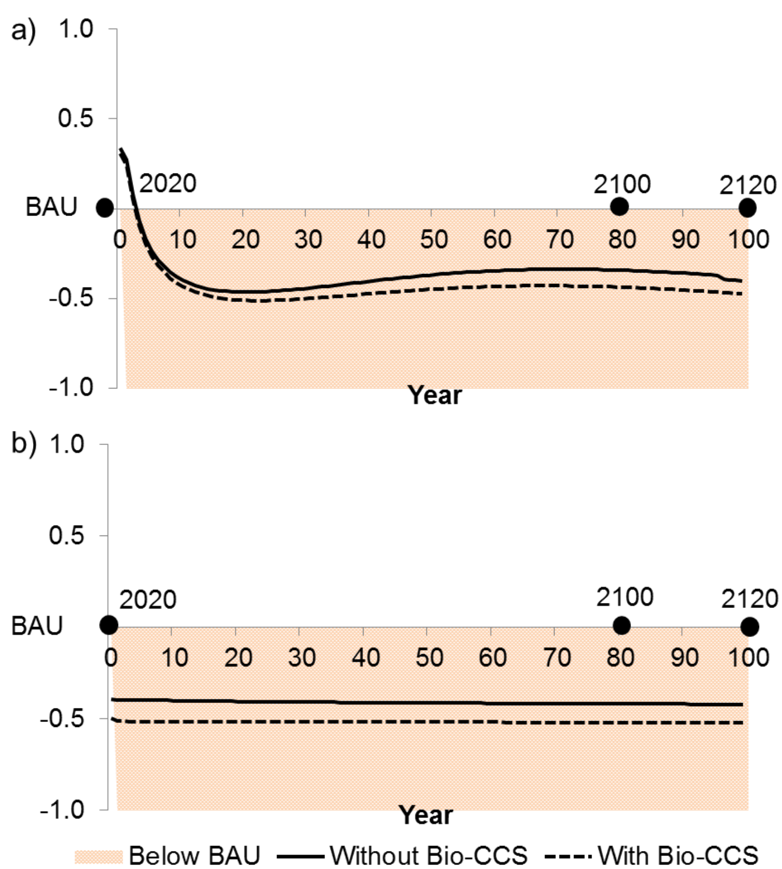
Figure 16 shows the CMI values for the production of electricity (1), biofuels and services (2) and biofuels, renewable-derived plastics and services (3) in scenario 2. The results improve from 1 to 3 according to the complexity of the mix of products achieving the best results when renewable-derived plastics are produced. In the case of Spain, in all cases there is a sharp reduction of the index from positive to negative and subsequent stabilisation except in the case of the electricity production (1) where the trend increase towards the climate worsening (from negative to positive) in the last years. In this case, the best configuration is the FBG with ICE and the worst the FBG with ORC option according to the efficiency. The highest efficiency, the highest climate mitigation. In all cases, the highest climate mitigation is achieved at a short time (first 20 years) since the transient emissions from the landfill are concentrated around 20 years after the landfilling of the MSW refuse. Considering Sweden (only 2 and 3), the climate change mitigation is obtained for the whole period considered, where an almost constantly mitigation is achieved. The mitigation for Sweden is higher than for Spain.

(1)



1.0
0.5
BAU
-0.5
-1.0
2020
2100
2120
0
10
20
30
40
50
60
70
80
90
100
Year
Below BAU
FBG/ICE
FBG/ORC
GG/SRC

(2)



a)
1.0
0.5
BAU
-0.5
-1.0
2020
2100
2120
0
10
20
30
40
50
60
70
80
90
100
Year
b)
1.0
0.5
BAU
-0.5
-1.0
2020
2100
2120
0
10
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30
40
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60
70
80
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100
Year
Below BAU
Without Bio-CCS
With Bio-CCS

(3)

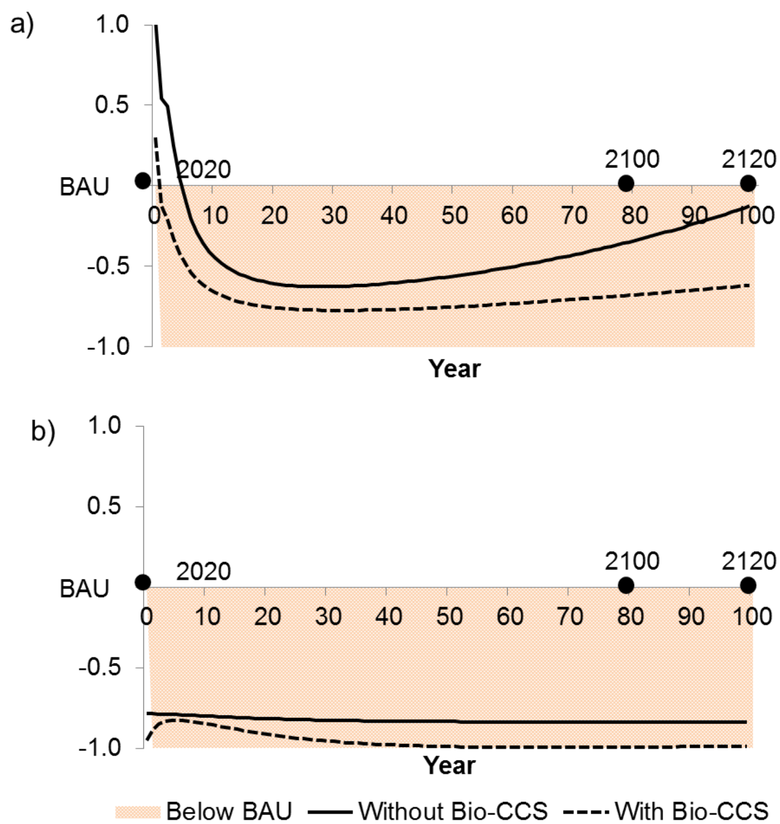


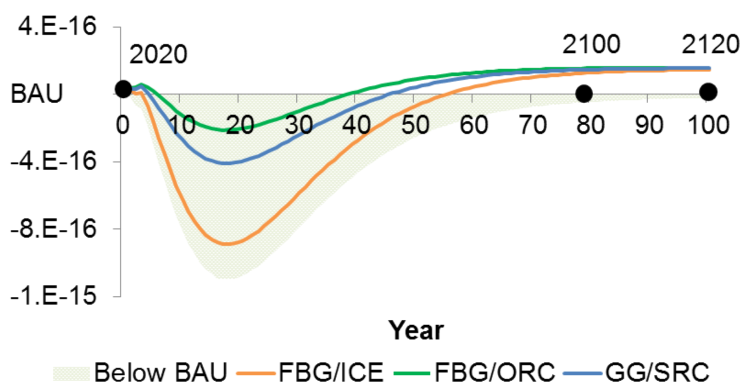
Figure 16. Climate mitigation index (CMI) for the production of electricity (1) and biofuels (2) and renewable-derived plastics (3) in Spain (a) and Sweden (b) in Scenario 2.

6.2.2. Differential climate impact (Paper III and IV)

Figure 17 shows the differential climate impact for the production of electricity (1), biofuels and services (2) and biofuels, renewable-derived plastics and services (3). The graphs 1 and 2 show the scenario 1 and the graph 3 the scenario 2 (only scenario 2 is assessed in Paper V, see Table 12). As CMI, the results improve from 1 to 3. Comparing these results with the static assessment, it is clear that these results could not have been predicted from the GHG saving or differential GHG impact. For instance, the differential GHG impact gave a higher climate benefit for Sweden. The static assessment gives an underestimation of the climate benefit in the production of biofuels in a landfill-dominant region like Spain. The results of incorporating Bio-CCS are also different from the stationary assessment. The effect of Bio-CCS incorporation is slightly lower in the time-dependent assessment.

In Figure 17-3, both the production of biofuels and the production of biofuels and renewable-derived plastics with Bio-CCS incorporation involve a climate benefit respecting to the current MSW refuse management scheme (BAU system) (Figure 17). In the case of Sweden the climate benefit is practically constant from the year 2030 whereas in the case of Spain, the climate benefit is higher in the first 40 years owing to the impact of the landfill emissions. The production of renewable-derived drop-in chemicals is a clear advantage regarding the production of biofuels in the Swedish case whereas in Spain this advantage is produced while the landfill emissions are being avoided.

(1)



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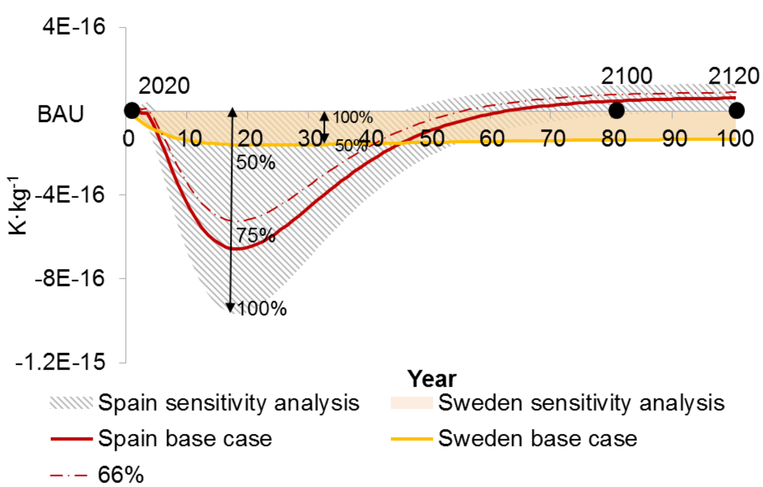

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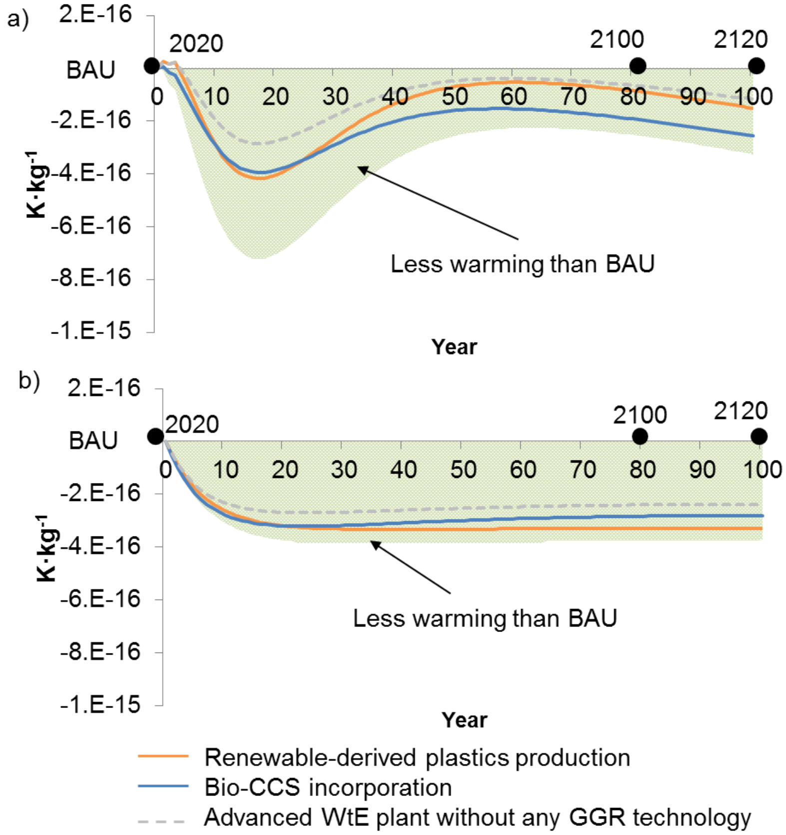

Figure 17. (1) Differential climate impact (DCI) for the production of electricity through different forms of production in Andalusia (South of Spain). (2) DCI (lines) and sensitivity to the biogenic fraction in MSW refuse (areas) for Spain (grey) and Sweden (brown) in scenario 1 producing biofuels and electricity. (3) DCI for the sustained production of renewable-derived plastics from MSW refuse, Bio-CCS incorporation to an advanced WtE plant producing transportation fuels and electricity and an advanced WtE plant producing transportation fuels and electricity without any GGR technology in Spain (a) and in Sweden (b) (scenario 2).

6.3. Techno-economic assessment (Paper V)

The economic comparison is shown in Figure 18, which displays the gate fee necessary to achieve 15% of internal return rate (IRR) as a function of plant fuel-input capacity (MWth), for the 4 WtE schemes considered. According to the previous discussion, incineration is considered for electricity loads higher than 100 MWth, whereas gasification schemes are displayed from small to medium capacities (5-60MWth) in order to adapt these WtE plants into existing MBT plants. As expected, the higher size plant allows lower gate fee to achieve the same economic feasibility. On the other hand, the impact of the gate fee on the feasibility is higher for gasification schemes as compared to incineration, since the reduction of the annualized cost of investment of these plants is more sensitive to the plant scale than incineration, increasing the cash flow more significantly than the wholesale electricity tariff. The FBG/ORC and FBG/IEA are seen to be better choices than GC/SRC from the economical point of view. The FBG/ORC configuration gives the lowest gate fee at a small-scale plant capacities (<35 MWth) because its lower capital and operational costs (Table 6). From 35 MWth, the FBG/ICE obtains the best results but the differences between the two technologies are very small, so it should be considered similar for the rough estimations considered in this work. However, ORC is a mature and widely spread and reliable form of energy production in CHP applications based on biomass (Turboden, 2016) with lower technical and economic requirements in gas cleaning than ICE option, so its suitability for market penetration in the short-term scenarios should be considered in terms of higher reliability.

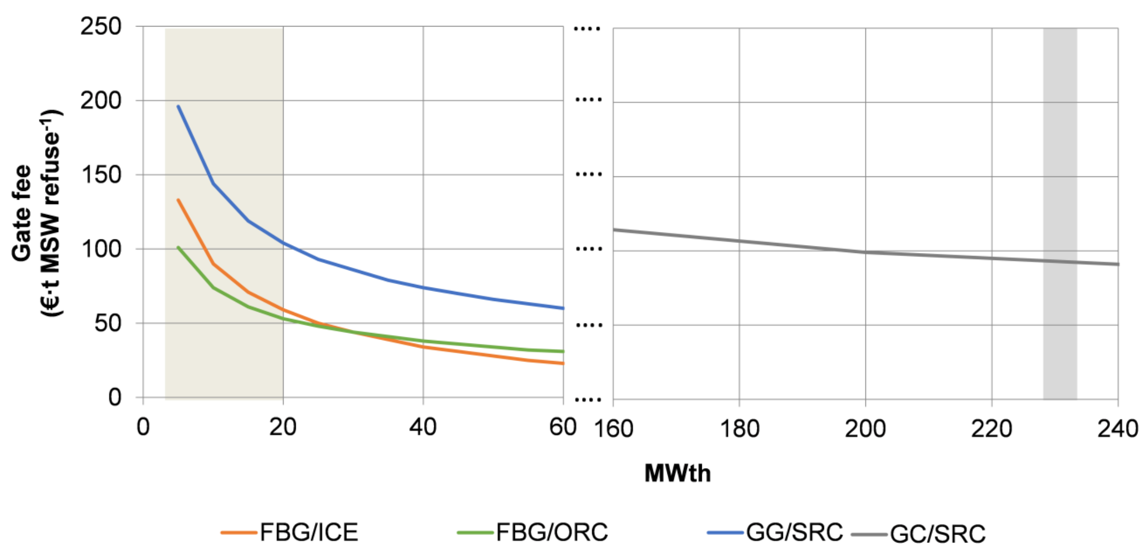

Figure 18. Economic results according to the WtE configuration (a. gasification, b. incineration) considering an IRR of 15% in Andalusia. The shaded area indicates the range of thermal energy in which the most of the MBT plants are (5-20 MWth) in Andalusia (a) and the incineration plant capacities proposed in this study (b).

7. Main findings of the thesis

- The impacts of using MSW refuse as a feedstock for the production of energy products are not well evaluated in the current European regulation. For instance, a saving of GHG emissions over 60% (according to the objective of the EU for 2018) could be achieved in advanced WtE plants producing biofuels and electricity from MSW refuse in a landfill-dominant country (Spain) only if Bio-CCS is incorporated. This result could be misleading with respect to the climate benefit in the avoidance of current MSW refuse management in both landfill- and incineration-dominant regions in Europe.
- The static GHG emission assessments, such as commonly found in LCA literature, cannot properly capture the impact of landfills on the evolution of GHG emissions with time and thus are unable to properly assess the climate change mitigation of the BIO system. Especially when considering potential dynamic evolution of both energy mix and MSW refuse management scheme in a 100-year period, only a time-dependent (dynamic) assessment can provide a proper impact assessment.
- Two parameters have been proposed for the dynamic GHG emission assessment: the climate mitigation index (CMI) and the differential climate impact (DCI). Both parameters have proven to be very effective and complementary in the evaluation of the climate benefit producing products and services from MSW refuse. Whereas the CMI allows a comparison of different disposal and conversion technologies for MSW refuse, it cannot reveal the actual impact on the average global temperature. Conversely, the DCI allows a quantification of the temperature change along with the climate impact of the biogenic carbon stored in GGR technologies (renewable-derived plastics, Bio-CCS or others) since the biogenic CO_2 emissions are excluded and carbon captured (in the form of plastic or CO_2) are counted.
- The promotion of landfill banning, despite being a very entrenched practice in Southern and Eastern Europe, would equalize the climate mitigation in the production of biofuels in Europe regardless their waste management scheme. This result is another example of how the contribution of landfills in climate change is still not fully understood. The estimated evolution of the waste management and electricity production for Spain and Sweden (as case examples) reveals that current landfill-dominant regions can even favorably compare with incineration-dominant regions thanks to the storage of biogenic carbon in the landfill.
- The production of renewable-derived plastics compares favorably in terms of climate benefit in the short and medium term with Bio-CCS 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 (IAMs) to include their global potential in forecasting scenarios to achieve the 2 ºC target.

- Both incineration and gasification-based WtE plants producing electricity in Southern Europe would allow a reduction of landfill disposal and GHG emissions meeting current European targets. The economics of their implementation is favored for gasification-based WtE plants compared with incineration plants.

8. Proposals for future research

The research conducted in this doctoral thesis has raised several aspects that are considered to be relevant for a future research. In this section, some of them, especially those related to the development of a sustainable waste management scheme, are highlighted.

- MSW is composed by different fractions, the biodegradable fraction of MSW or biowaste is one of them. Biowaste is the fraction composed by food scraps, gardening and pruning residues, cellulose and wood. This fraction is currently separated at source or in a MBT plant and used for composting and/or anaerobic digestion. Composting is considered a type of recycling; therefore, it fulfills the waste hierarchy since recycling must be prioritized against valorization. However, the Waste Directive establishes waste hierarchy can be broken from a life cycle thinking. Because of that, anaerobic digestion is also very common in the biowaste management and; in the same way, advanced WtE plants could use biowaste as feedstock. The advantage of biowaste is their consideration as pure biomass according to RED.
- Landfilling has been being a common practice in Europe, therefore, numerous closed landfills can be found in all the European countries. A lot of these landfills sites were closured before the recycling promotion, so they contain many recyclable materials. Enhanced landfill mining (ELFM) is a process whereby solid wastes, which have previously been landfilled, are excavated and processed. It enables the recovery of valuable materials that can be brought back into the cycle and also allows for recovering land area. Recently, European Parliament has decided to include a specific reference to "Enhanced Landfill Mining" in Landfill Directive (EURELCO, 2017) proposing a regulatory framework for it to permit the retrieval of these secondary raw

materials that are present in existing landfills. Therefore, the assessment of the climate mitigation of landfill mining must be carried out. A combination of material and energy recovery where the excavated waste can be directly recycled or pre-treated and used in the RDF production to feed an advanced WtE plant (gasification-based) is proposed. Both material and energy recovery contribute to a circular economy using MSW refuse as a secondary raw material.

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_{mix}	emission factor from electricity grid mix (g CO_2 eq.·MJ^{-1})
$e_{landfill}$	biogenic carbon storage in landfilled plastics (g CO_2 eq.·MJ^{-1})
e_{pool}	biogenic carbon storage in plastics (g CO_2 eq.·MJ^{-1})
e_p	emissions from processing (g CO_2 eq.·MJ^{-1})
e_{pt}	emissions from pretreatment (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_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	ratio of chemical production (1 BAU system, 2 BIO system) (%)
x_i	fraction of heat production (1 BAU system; 2 biorefinery, 3 heat mix in BIO system) (%)
y_i	ratio of electricity production (1 landfill, 2 incineration in BAU system; 3 electricity grid, 4 biorefinery In BIO system) (%)
z_i	ratio of biofuel production (1 fossil fuels in BAU system; 2 biorefinery in BIO system) (%)

Greek letters

α	Biogenic fraction of MSW refuse (%)
β	Anthropogenic emission factor from MSW refuse incineration (%)
$\eta_{conversion}$	Conversion efficiency from chemical to plastic (%)

Abbreviations

AGTP	Absolute Global surface Temperature change Potential
AGWP	Absolute Global Warming Potential
e	Carbon flows
BAU	Business as usual
BIO	Bioenergy
BECCS/Bio-CCS	Carbon capture and storage in bioenergy
DCI	Differential climate impact
DME	Dimethyl ether
$E_{BIO/BAU}$	GHG impact of the BIO or BAU system
E_{TB}	Global GHG balance of the thermochemical biorefinery
EF	Emission factor (fossil comparator)
EPA	U.S. Environmental Protection Agency
FBG/ICE	Fluidised bed gasifier/internal combustion engine
FBG/ORC	Fluidised bed gasifier/ organic Rankine cycle
GC/SRC	Grate combustor/steam Rankine cycle
GG/SRC	Grate gasifier/steam Rankine Cycle
GGR	Greenhouse gases removal
GHG	Greenhouse gases
IPCC	Intergovernmental panel on climate change
LCA	Life cycle assessment
LHV	Lower heating value
MSW	Municipal solid waste
P	Products still in use
PVC	Poly vinyl chloride
RDF	Refuse derived fuel
SM	Supplementary material
W	Waste
WMGHG	Well mixed GHG

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Paper I

GHG Saving in Thermochemical Biorefineries Using Municipal Solid Waste in Andalusia

This paper presents the static GHG emission assessment of three configurations of a thermochemical biorefinery: a conventional thermochemical biorefinery producing biofuels (ethanol and dimethyl ether: DME) and electricity, a thermochemical biorefinery with multiproduction producing chemicals (methyl acetate) and a thermochemical biorefinery incorporating Bio-CCS (bioenergy with carbon capture and storage). It has been published as proceedings in 2014[12].

1. Introduction

In Andalusia, millions of tonnes of municipal solid waste (MSW) are generated every year and only about a half of them are recycled or composted, the rest ends up in the landfills (waste incineration is not allowed in Andalusia). However, this waste has an acceptable heat value (around 16 MJ/kg) that should be used minimizing the waste volume, decreasing the negative impact of landfills, avoiding depletion of fossil fuels and reducing greenhouse gases (GHG) emissions to the atmosphere.

A thermochemical biorefinery is a facility that processes biomass by means of gasification to produce biofuels, chemicals and electricity. The use of MSW as feedstock in this kind of biorefinery (Figure 1) has the advantage of removing waste that nowadays goes to landfill disposal. In addition, the waste is available throughout the year and currently free of charge. Moreover, MSW has a 50% renewable fraction according to International Energy Agency (IEA, 2012).

GHG emissions are reduced in biorefineries based on MSW since there are not emissions associated with harvest or direct and indirect land-use change, only with the MSW

[12] Aracil C, Haro P, Ollero P, Vidal-Barrero F. GHG saving in thermochemical biorefineries using municipal solid waste in Andalusia. EUBCE 2014, pp 1576-1578 http://www.etaflorence.it/proceedings/?conference=2014&mode=author&letters=a&items=Aracil%2C+C.&categories=0

transport to the biorefinery. Even these emissions can be avoided if the biorefinery is next to the landfill. This is the assumption made in this study.

This study is based on a set of twelve configurations (process concepts) of thermochemical biorefineries previously published (Haro el at., 2013) that have been classified into three categories: conventional biorefinery (A), biorefinery with multiproduction (B) and biorefinery with Bio-CCS (C).

Figure 1. Visual concept of the study.

2. Objectives

- Assessment of GHG emissions of a plant of 50 MW based on municipal solid waste refuse (organic and non-recyclable fraction) with possibility of Bio-CCS (Bioenergy with Carbon Capture and Storage) incorporation.
- Assessment of the impact of co-production of chemicals (non-energy drivers) in the GHG balance of thermochemical biorefineries (multiproduction).
- Assessment of the impact of Bio-CCS incorporation in the GHG balance of thermochemical biorefineries.

3. Methodology

Directive 2009/28/EC (Directive 2009/28/EC) gives a general formula for the calculation of GHG emissions of biofuels and bioliquids. This formula considers neither MSW like feedstock nor chemicals like potential products, so a modified and/or extended (Figure 2) formula is used in this work. Moreover, the value of emissions in the final use of biofuels is defined as zero in the Directive (neutral impact of biofuels). However, this is a simplification and we have considered the carbon embodied in the non-renewable MSW fraction and the fraction of other gases with global warming potential (GWP) like N_2O and CH_4.

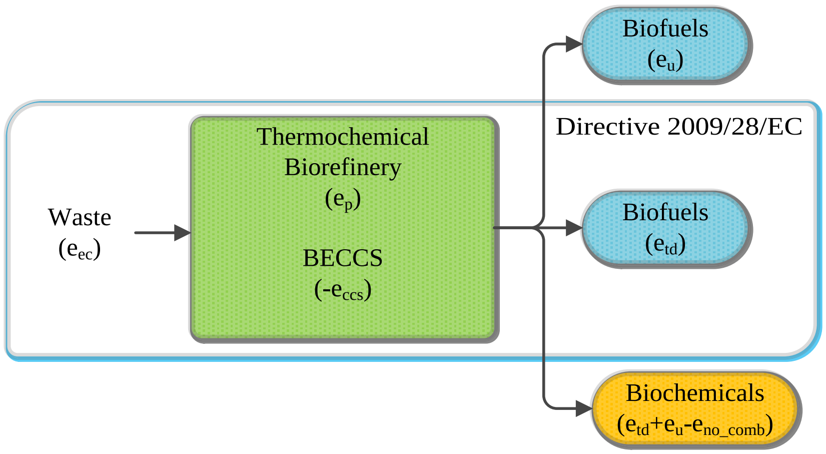

Figure 2. Considered emissions in the GHG calculation.

4. Results

4.1. Conventional biorefinery

The first part of the assessment consists in the calculation of GHG saving in a conventional biorefinery that only produces biofuels and electricity. We have considered four configurations depending on the mix of final products and syngas conditioning (identified by the reformer) (Table 1).

Although the results are discouraging (Figure 3), the best one is obtained in the configuration A1 where ethanol and electricity are produced and a steam reformer (SR) is used. In this case, GHG saving is just over 20%. The cases A3 y A4 use autothermal reformer (ATR) and consume oxygen in the gasifier thereby increasing GHG emissions because of the energy consumption of the air separation unit.

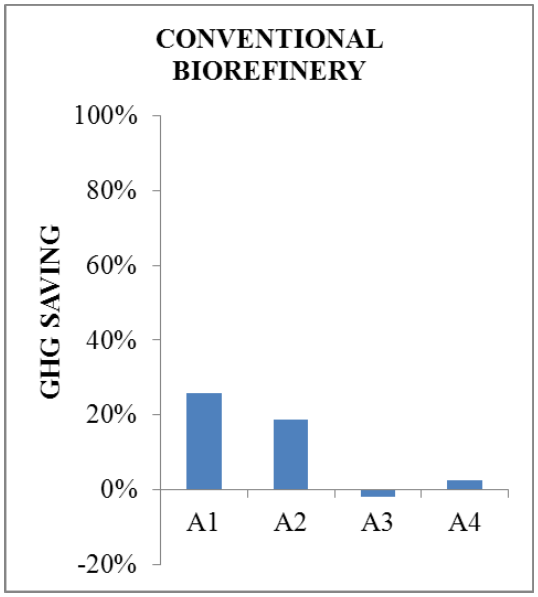

Figure 3. GHG saving of conventional biorefinery producing biofuels and electricity (A).

Table 1. Configurations of conventional biorefinery (A).

	CONVENTIONAL BIOREFINERY (A)			
	A1	**A2**	**A3**	**A4**
Ethanol	✓	✓	✓	✓
DME		✓		✓
Methyl acetate				
Hydrogen				
Electricity	✓	✓	✓	✓
Reformer	SR	SR	ATR	ATR

4.2. Biorefinery with multiproduction

The second part of the assessment consists in the calculation of the GHG saving in a thermochemical biorefinery with multiproduction producing biofuels, chemicals and electricity. We have considered four configurations of the biorefinery depending on the mix of final products and the used reformer in the plant (Table 2).

The results improve considerably due to the chemical co-production (Figure 4). Even three of the configurations exceed the 60% reduction threshold (target for 2018 in EU). The worst result is for B1 since it uses an ATR as well as A3 and A4.

The benefit of chemical co-production relies on the negative value of the emissions in their final use since they are not combusted. Chemicals represent a net storage of carbon for a period of time.

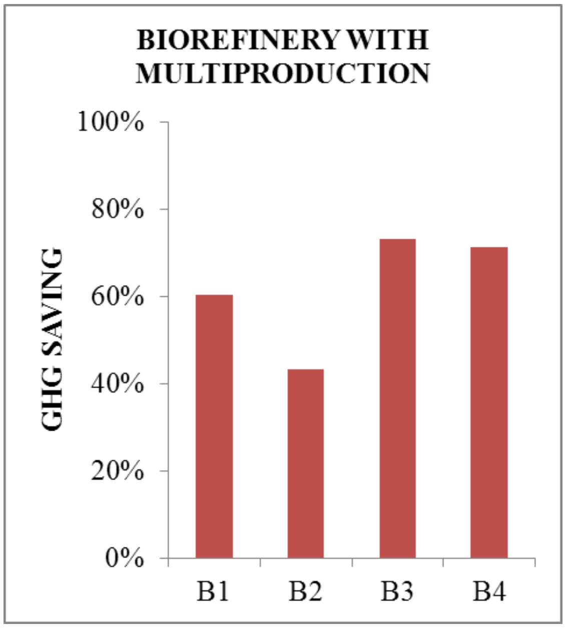

Figure 4. GHG saving of biorefinery with multiproduction producing biofuels, chemicals and electricity (B).

Table 2. Configurations of biorefinery with multiproduction (B).

	BIOREFINERY WITH MULTIPRODUCTION (B)			
	B1	**B2**	**B3**	**B4**
Ethanol				
DME	✓	✓	✓	✓
Methyl acetate	✓	✓	✓	✓
Hydrogen			✓	
Electricity	✓	✓		✓
Reformer	SR	ATR	TR	TR

4.3. Biorefinery with Bio-CCS

The third part of the assessment consists in the calculation of GHG saving in a thermochemical biorefinery incorporating Bio-CCS. We have considered four configuration of the biorefinery depending on the mix of final products (Table 3).

The results improve drastically (Figure 5). All cases use the same reformer (Tar reformer, TR) and are over 90% saving of GHG emissions.

The CO_2 sequestrated via Bio-CCS are considered as a negative contribution in the GHG balance, since they constitute a net outlet of CO_2 from the atmosphere if renewable fraction is considered or avoided emissions if not.

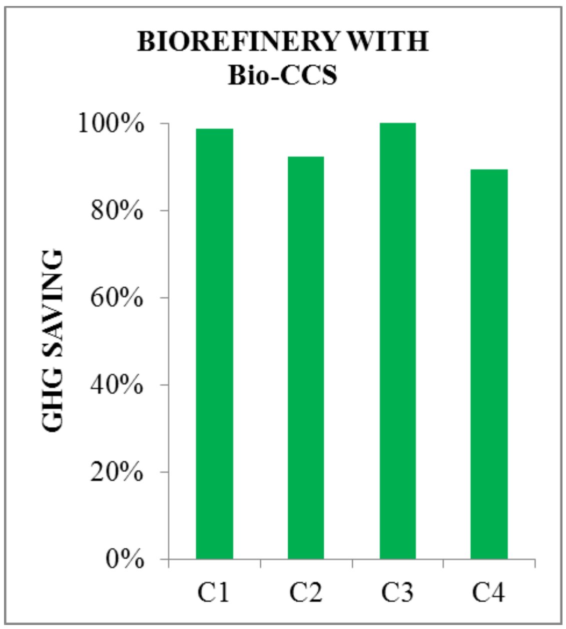

Figure 5. GHG saving of thermochemical biorefinery incorporating Bio-CCS (C).

Table 3. Configurations of biorefinery with Bio-CCS (C).

	BIOREFINERY WITH Bio-CCS (C)			
	C1	C2	C3	C4
Ethanol	✓	✓	✓	✓
DME			✓	✓
Methyl acetate				
Hydrogen		✓		✓
Electricity	✓	✓	✓	
Reformer	TR	TR	TR	TR

5. Conclusions

GHG saving over 60% (target for 2018 in EU) could be achieved in thermochemical biorefinery based on MSW with multiproduction and/or Bio-CCS incorporation. Therefore, MSW are an alternative feedstock to energy crops for thermochemical biorefineries. In addition, the fact that the waste is free or even its management can be charged (extra income), allows more affordable plant sizes (50 MW).

Although thermochemical biorefineries based on MSW can hardly contribute to the achievement of the European Union objectives in 2020 because of their status, it could play an important role for a long-term sustainability in Europe.

Nomenclature

ATR	autothermal reformer
Bio-CCS	bioenergy with carbon capture and storage
CCS	carbon capture and storage
DME	dimethyl ether
e_{ccs}	emissions saving from carbon capture and geological storage
e_{ec}	emissions from the extraction or cultivation of raw materials
$e_{no\text{-}comb}$	negative emissions because of no combustion of chemicals
e_p	emissions from processing
e_{td}	emissions from transport and distribution
e_u	emissions from the final use
GHG	greenhouse gas
GWP	global warming potential
MSW	Municipal solid waste
SR	Steam reformer
TR	Tar reformer

Acknowledgements

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Haro P, Ollero P, Villanueva Perales AL and Gómez-Barea A. Thermochemical biorefinery based on dimethyl ether as intermediate: Technoeconomic assessment. Apply Energy 2013;102: 950-61.

Paper II

Time-integrated GHG emissions in a thermochemical biorefinery producing ethanol from MSW in Andalusia

This paper presents the assessment of the time-integrated GHG emissions of a thermochemical biorefinery producing ethanol and electricity. It has been published as conference proceedings in 2015[13].

1. Introduction

Andalusia is the most populated and the second largest region of Spain occupying 90.000 km^2. Every year, five millions tons of municipal solid waste (MSW) are generated and almost the 80% ends up in the landfills (Regional non-dangerous residues management Director Plan of Andalusia 2010-2019). The waste management in Andalusia is based on sorting plants, recycling plants and landfills; there are no waste-to-energy facilities. Landfilling is an environmental problem that can be solved at the same that renewable energy is produced since European Commission encourages the use of residues and waste to produce bioenergy (Report on the proposal for a directive of the European Parliament, 2012). Because of that we propose an energy recovery from MSW in Andalusia saving GHG emissions, minimizing landfill disposal and avoiding the negative impact of landfills in the soil, the groundwater and biodiversity.

2. Our proposal

2.1. A new concept of thermochemical biorefinery

We propose the use of the non-compostable and non-recyclable fraction of the MSW (refuse) as feedstock in a thermochemical biorefinery producing an energy carrier (ethanol) and electricity by means of gasification. First of all, the refuse has to be

[13] Aracil C, Haro P, Ollero P. Time-integrated GHG emissions in a thermochemical biorefinery producing ethanol from MSW in Andalusia. EUBCE 2015, pp 1376-1379. http://www.etaflorence.it/proceedings/?conference=2015&mode=author&letters=a&items=Aracil%2C+C.&categories=0

converted into refuse derived fuel (RDF) in a process based on size reduction, inerts separation, drying and pelletization. This process increases the heat value and the density of the fuel improving its quality and making it more manageable. The RDF would be used to feed a 100 MWth thermochemical biorefinery producing ethanol and electricity for 20 years (Table 1). The final products and the plant lifetime have been chosen considering the current and the future characteristics of the energy system in Europe but other possibilities can be assessed. The chemical route of the thermochemical biorefinery has been previously published (Haro et al.,2013). In this study twelve configurations of thermochemical biorefinery were techno-economically assessed using poplar chips as feedstock. We choose one of the process concepts according to the techno-economic results (i.e. TAR-01).

Three sorting plants have been chosen in Seville, the capital city of Andalusia, since they produce enough amount of MSW refuse to feed the thermochemical biorefinery. Figure 1 shows the location of the three sorting plants and the thermochemical biorefinery.

Figure 1. Location of the three sorting plants selected to provide the feedstock to the thermochemical biorefinery in Seville (Andalusia, Spain).

Table 1. Technical data about the thermochemical biorefinery.

Plant capacity (MWth)	Total energy efficiency	Ethanol production (t/h)	Electricity production (GWe)
100	43.6%	4	100

2.2. GHG balance

According to Renewable Energy Directive (RED), we carried out the stationary assessment of the GHG emissions called GHG balance (Directive 2009/28/EC). MSW is not considered in RED, so an adapted methodology we previously published was used (Haro et al., 2015).

GHG balance is calculated for two scenarios, the reference scenario corresponding to the landfill and the use of fossil fuels to produce the same mix of products; and the bioenergy system. The fossil reference for transportation fuels is given in RED (83.8 g CO_2/MJ). Although more recent studies indicate that a higher number would be more accurate (Commission staff working document, 2012), we assume the value of the current directive.

The landfill has been comprehensive assessed since there is scarce information about Andalusian landfills. A typical bin in Andalusia contains a 40-50% of organic matter, food scraps mainly. This organic matter is recovered and composted. After composting, the organic matter content in the refuse is still predominant. Because of that, the production of biogas in the Mediterranean landfills is so high that biogas is recovered and used to produce electricity or to burn in a flare. However, the recovery is not very efficient and a 30-50% of the biogas is unrecovered and emitted to the atmosphere as CO_2 and CH_4. Moreover, the biogas combustion also produces non-biogenic CO_2 emissions. Considering that and the parameters that Intergovernmental Panel on Climate Change (IPCC) and Environmental Protection Agency (EPA) published in 2014 and 2006 respectively, we obtain a result of 64.4 g CO_2/MJ (Figure 2) (Pipatti et al., 2006, EPA, 2014).

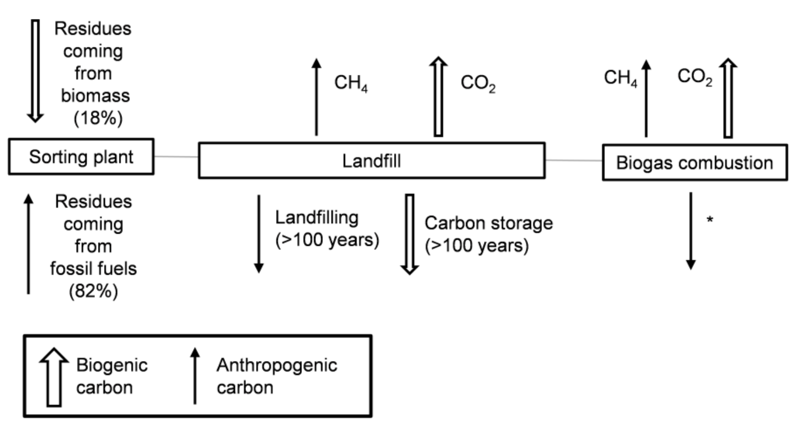

Figure 2. GHG balance in a landfill.

The bioenergy system is also evaluated considering the emissions associated to feedstock transport, ethanol distribution, emissions factors of the chemicals and commodities used in the thermochemical biorefinery and flue gas; and finally, ethanol combustion. The result is a GHG balance of 26.2 g CO_2 eq/MJ.

According to RED, the CO_2 emissions from the biogenic fraction of MSW has not been considered in the calculations, only the GHG emissions from de fossil fraction and the non-CO_2 GHG emissions from the biogenic fraction (Figure 3).

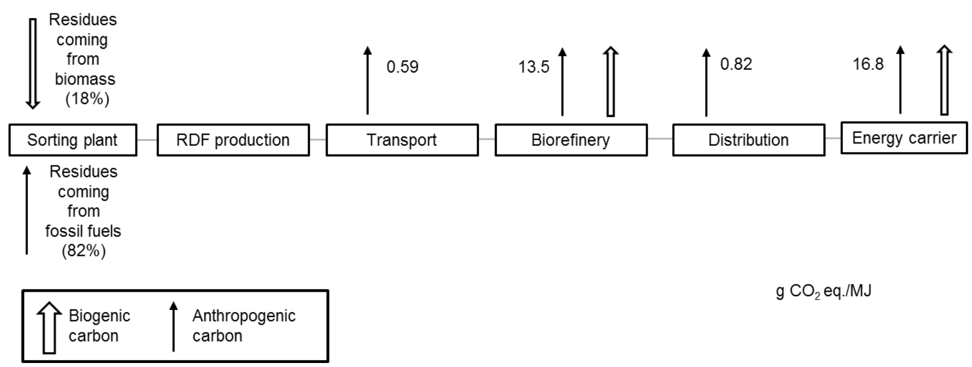

Figure 3. GHG balance in the BIO system.

Finally, at the end of the GHG balance calculations we have a value for every system. The bioenergy system and the fossil reference are represented by a plant producing the same mix of products, but in the bioenergy system the feedstock is the RDF from MSW and in the fossil reference is fossil fuels. Then, the value obtained in the GHG balance is constant during the 20-year lifetime. However, the landfill reference is completely different; the result of the GHG balance is an average. The way in which the biodegradable fraction of the MSW is degraded is slow and no constant. During 40 years more or less, a ton of MSW disposed in the landfill in year 0 can produce emissions, the first and the last years the emissions are lower and in the intermediate years the emissions are higher and also emitted GHG are different in each step. Figure 4 shows the GHG emissions distribution of each system per year.

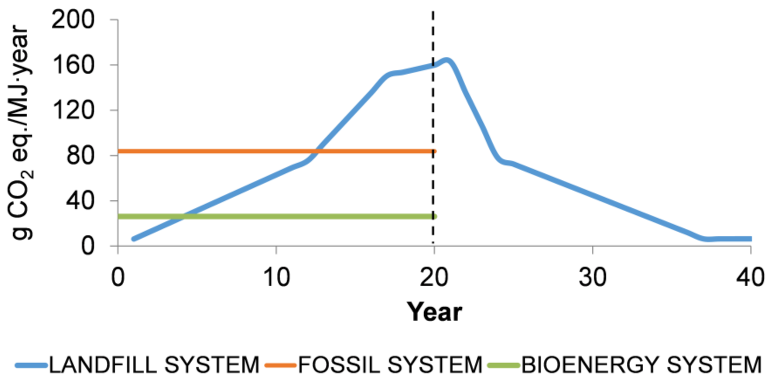

Figure 4. GHG emissions per year in the three assessed systems.

3. Time integrated GHG emissions

After the GHG balance, we are ready to assess the impact of these GHG emissions in the atmosphere.

The assessment of the time-integrated GHG emissions is the newest and most important part of this study. This part consists in the integration of the natural dynamics of the atmosphere using Pulse emission (PE) and Radiative Forcing (RF) according to IPCC (Myrhe et al., 2013, Solomon et al., 2007). The objective is the determination of the time horizon in which GHG emissions have an impact in the atmosphere carbon balance.

3.1. Methodology

The pulse emission is the sum of all the emissions produced during every year. We use the GHG balance previously calculated as pulse emissions (Figure 4). Then, the first step is the calculation of the CO_2 remaining in the atmosphere per year by multiplying the GHG balance by the natural decay of the pulse emission (Equation 1).

$$PE(t) = a_0 + \sum_{i=1}^{3} a_i * e^{-t/\tau_i} \tag{1}$$

where

PE: natural decay of pulse emissions.

t: year in which emissions are produced

The Table II shows the values of the rest of parameters.

Table 2. Values of the pulse emissions parameters.

a_0	a_1	a_2	a_3	τ_1	τ_2	τ_3
0.217	0.259	0.338	0.186	172.9	18.51	1.186

The second step is the calculation of the cumulative CO_2 as grams of CO_2 equivalent per mega joule of output and that as CO_2 concentration in the atmosphere in ppmv.

The CO_2 concentration is used to calculate instantaneous radiative forcing (Equation 2) and then cumulative radiative forcing multiplying by the seconds in a year (Sathre et al., 2011).

$$IRF = \frac{3.7}{ln2} * \ln(1 + \frac{[CO_2]}{C_0}) \quad (2)$$

where

IRF: Instantaneous Radiative Forcing (W/m^2)

$[CO_2]$: change in the CO_2 concentration in the atmosphere

C_0: CO_2 concentration in the atmosphere

4. Results

4.1. Cumulative CO_2

Cumulative CO_2 measures the grams of equivalent CO_2 per mega joule of output accumulated in the atmosphere during 100 years contributing to the intensification of the greenhouse effect. In year 20, the thermochemical biorefinery stops the production, that implies the end of the emissions, because of that the CO_2 accumulation in the atmosphere starts to decrease. This situation is identical in the fossil reference (a petrochemical refinery producing the same mix of products). However, the situation is different in the landfill, the disposal stops in year 20 but not the emissions because the waste continues degrading for 20 years more producing and emitting biogas. The Figure 5 shows the turnaround in the year 20.

At the end of the proposed time horizon (100 years), there is CO_2 remaining in the atmosphere in the three systems, although it is five times higher in the case of the landfill than in the bioenergy system (Figure 6).

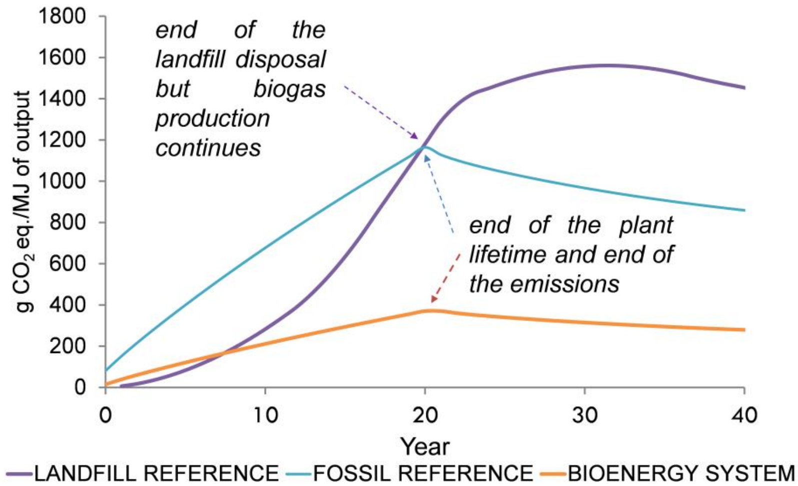

Figure 5. Time-integrated GHG emissions of the three studied systems for 40 years.

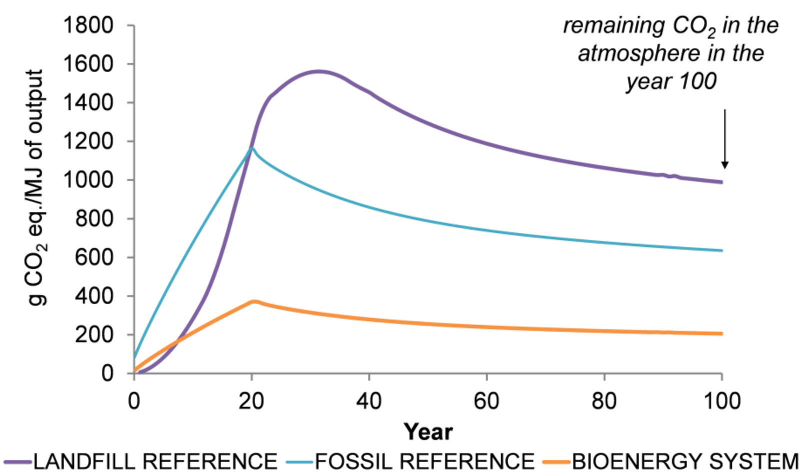

Figure 6. Time- integrated GHG emissions of the three studied systems for 100 years.

4.2. Cumulative Radiative Forcing (CRF)

Radiative Forcing is a measure of the GHG induced imbalance between incoming and outgoing radiation in the earth system (W/m^2). When Radiative Forcing is multiplied by the seconds of a year, Cumulative Radiative Forcing (CRF) is obtained ($W{\cdot}s/m^2$). Figure 6 shows the results of this parameter for the reference system (landfill and fossil reference) and bioenergy system. The area between the curves represents net emissions. In the case of using MSW as feedstock the net emissions are avoided, then a lower global warming is produced.

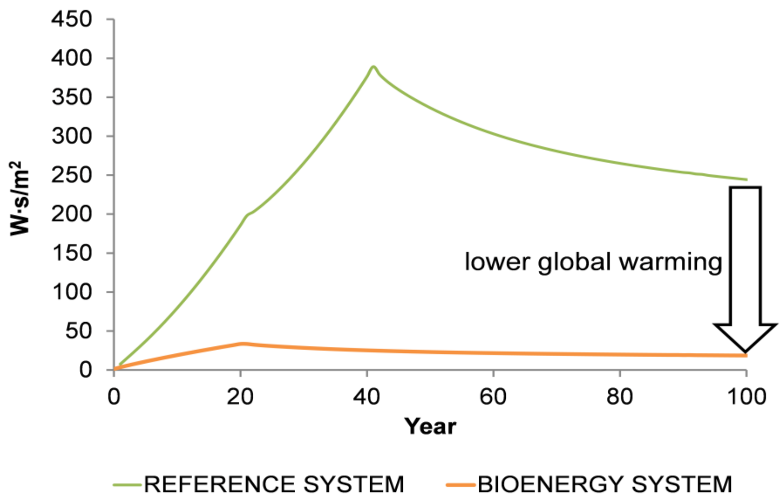

Figure 7. CRF of the two scenarios. The area between the curves represents net emissions to the atmosphere.

5. Conclusions

MSW landfilled degrades producing emissions for 40 years or more, their impact in the atmosphere is still considerable in the year 100 (1 kg CO_2 eq./MJ of output) and higher than those of the fossil and bioenergy system.

Our proposal of assessment of time-integrated GHG emissions allows to assess the size of the impact of a energy system in the atmosphere, particularly important in the case of MSW landfills where the emissions are slowly produced.

MSW as feedstock can play an important role in the future of the bioenergy saving GHG emissions, minimizing landfill disposal in regions as Andalusia.

Acknowledgements

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Paper III

Proving the climate benefit in the production of biofuels from municipal solid waste refuse in Europe

This paper aims to properly assess the impact of the production of biofuels from MSW refuse on climate change by applying several methodological improvements in a time-dependent assessment, i.e., an explicit consideration of biogenic carbon flows using a dynamic LCA and an absolute formulation of the cumulative and instantaneous climate metrics. It has been published as paper in 2017[14].

1. Introduction

The impact of anthropogenic greenhouse gases (GHG) emissions on climate change is a subject of growing public concern. From 1972 to the present, numerous Climate Change Summits have joined scientists from nations around the world to analyse the increasing concentration of CO_2 in the atmosphere and its consequences on the climate. The Kyoto Protocol was the first international agreement aiming at curbing GHG emissions, but its implementation has been only partially successful. The first-ever universal, legally binding global climate post-Kyoto deal has been adopted in Paris in December 2015. This agreement relies on pledges from signatory countries to drastically reduce GHG emissions from transport and industry sectors by 2030 in order to maintain the temperature anomaly below 2 °C, or even 1.5 °C, compared to the pre-industrial period (Adoption of the Paris Agreement, 2015).

The International Energy Agency (IEA) stated that the world cannot emit more than around 1000 Gt of CO_2 from 2011 onwards in order to achieve a 2 °C target (ETP, 2012, WEO, 2014). According to the IEA *BLUE map scenario*, 3,000 Mtoe of biomass and waste (8% of the energy mix for 2050) will be demanded as primary energy to achieve this objective (ETP, 2010). However, the IEA claims that, along with bioenergy production, it will also be

[14] Aracil C, Haro P, Giuntoli J, Ollero P. Proving the climate benefit in the production of biofuels from municipal solid waste refuse in Europe. J Clean Prod 2017;142:2887-2900.
Article and Supplementary data in https://doi.org/10.1016/j.jclepro.2016.10.181

crucial to incorporate negative-carbon technologies, such as biogenic carbon capture and storage (Bio-CCS) (ETP, 2010). Bio-CCS is said to have a potential for carbon abatement ranging from 3 to 10 Gt of CO_2 equivalent per year according to Fifth Assessment Report of the International Panel of Climate Change (IPPC, 2014, Lomax et al., 2015a).

In Europe, the Renewable Energy Directive (RED) incentivises the production of bioenergy from different types of biomass sources allowing the EU Member States to support biofuels production, e.g. tax exemptions or quotas (Directive 2009/28/EC). However, the use of food crops for biofuel production has created a controversy since GHG emissions linked to indirect land-use change have been shown to be significant, in many cases actually making biofuels more GHG intensive than fossil fuels (ILUC, 2015, Lapola et al., 2010). Consequently, the recent RED amendment imposes a cap on the use of food crops and clearly promotes the use of waste and residue feedstocks (Directive 2015/1513). Nonetheless, potential environmental risks associated to biofuels production from wastes and residues have been raised in the literature (Giuntoli et al., 2015, Niziolec et al., 2015, Onel et al., 2014, Rada, 2014, Samer, 2015, Schmitt et al., 2012, Stichnothe and Azapagic, 2009, Wang et al., 2012,). Regarding GHG emissions, literature is still scarce (Niziolec et al., 2015, Onel et al., 2014, Schmitt et al., 2012, Stichnothe and Azapagic, 2009, Wang et al., 2012).

Municipal Solid Waste (MSW) is one of the waste materials considered as a possible source for biofuels production in both the RED and the literature. The use of MSW in production processes is in agreement with the principles of the Circular Economy, where fossil fuel extraction and waste generation are their key drivers. MSW also compares favourably with other waste and residue feedstocks since it is available throughout the year, it is concentrated (supply locations), and it is costless or even a direct source of revenues due to the negative cost paid for its disposal, e.g. landfill gate fee (Arena et al., 2015, Manfredi et al., 2015). MSW is a heterogeneous mixture of different waste materials, such as food scraps, plastics, paper and cardboard, wood, textiles and inert materials. The composition of MSW depends on the waste management system, feeding habits and economic development of the region considered (Rada, 2014). In accordance with the European waste hierarchy, only the non-recyclable fraction of MSW, called MSW refuse, can be directly used for energy recovery, including electricity and fuel production; while the use of other waste fractions must be justified by a life-cycle thinking (Directive

2008/98/EC). The MSW refuse can be identified in Europe by the codes shown in Table S1 of the Supplementary Material (SM) (Commission Decision 2000/532/EC).

In 2013, 242 million tons of MSW were generated in Europe and the 57% of them, i.e., the MSW refuse, were incinerated or landfilled (Eurostat). Considering a lower heating value between 8 and 12 MJ/kg (Boesch et al., 2014, Consonni and Viganò, 2012, Yassin et al., 2009), the MSW refuse generated in Europe would be equivalent to 1,250 PJ/year. Therefore, MSW refuse is an energy source similar to agricultural residues in Europe (Biomass futures). However, as it has been proved for first generation biofuels (indirect land-use change) (Directive 2009/28/EC), it is mandatory to avoid a shifting of environmental burdens if a change in MSW refuse management is going to be promoted.

Currently, MSW refuse is either disposed in landfills or incinerated, in both cases with partial energy recovery. However, the situation is not even throughout the continent. In Northern and Central Europe, MSW refuse is mainly incinerated with energy recovery in waste-to-energy plants and landfill disposal is limited or even banned whereas in Southern and Eastern Europe, the MSW refuse is mainly landfilled with partial biogas recovery and used for electricity production, and only to a lesser extent incinerated (Figure 1) (Eurostat).

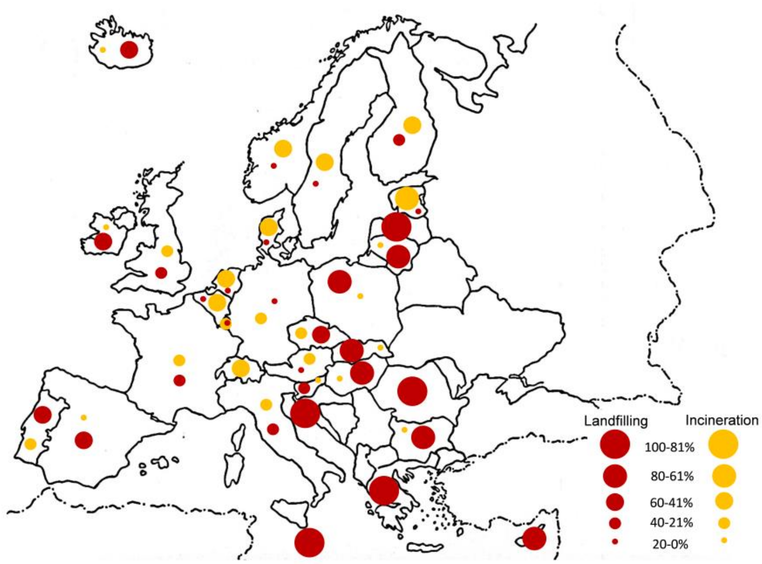

Figure 1. Landfilling and incineration ratios of MSW refuse in Europe (elaborated from Eurostat).

In a thermochemical biorefinery producing biofuels from MSW refuse, the Refuse Derived Fuel (RDF) can be produced and processed with the same processing technologies used for the production of biofuels from lignocellulosic biomass: pyrolysis and gasification. Considering RDF, gasification offers a higher adaptability and versatility (it allows the co-production of fuels and electricity) (Adefeso et al., 2015, Couto et al., 2015, Niziolec et al., 2015, Onel et al., 2014,). The main drawback is the technical limitations of syngas cleaning (e.g. removal of tars, heavy metals and inorganic compounds) (Panepinto et al., 2014, Prabhansu et al., 2015). Another option is to consider a biochemical biorefinery for the production of biofuels from MSW refuse. The heterogeneity of MSW refuse and the high concentration of heavy metals make their biochemical conversion difficult (Ballesteros et al., 2010, Mu et al., 2010) and therefore, this option is not considered as feasible for biofuel production. However, biochemical conversion is usually associated to the organic sorted fraction of MSW (Ballesteros et al., 2010, Colón et al., 2012).

2. Goal and scope of the study

Despite the support to MSW refuse for the production of biofuels, to the authors' knowledge, a comprehensive study analysing the actual climate benefit from the shift of MSW refuse management system in different European regions is still missing. This study aims to cover this gap by analysing the climate benefit in the production of biofuels from MSW refuse in Europe looking for possible climate burdens.

Firstly, the emissions associated to the different MSW refuse management alternatives (landfilling, incineration and production of biofuels) are assessed separately. Then, we calculate the GHG balance, saving and differential impact of biofuels production as indicated in our previous studies based on European regulation (static assessment). We analyse two EU member states (Spain and Sweden), considering two scenarios: one in which the current MSW management has been extended, unchanged, for the next 100 years, and one where it varies to follow EU Directives (phasing landfill disposal out) together with an evolution of emissions from the electricity mix and transportation fuels.

Because the release of GHG emissions in landfills is dynamic, time-dependent parameters are necessary to provide a complete impact assessment. We assess two combinations: the climate change mitigation potential of different MSW refuse management options and the climate change mitigation achievable in the two member states.

Finally, we test the sensitivity to the main parameters in the study, such as biofuel production efficiency, biogas collection efficiency in the landfill, fraction of carbon captured by Bio-CCS and biogenic fraction in the MSW refuse.

3. Materials and methods

The recent debate concerning the proper assessment of the climate impact of bioenergy has highlighted that attributional Life Cycle Assessment (LCA) studies can incur significant shortfalls (Plevin et al., 2014). The latest LCA literature on bioenergy systems has seen the implementation of methodological improvements to provide a more complete impact assessment (Giuntoli et al., 2015), which are used in this study.

Firstly, ignoring biogenic-CO_2 flows and considering biomass as inherently and instantaneously carbon neutral lead to erroneous results (Agostini et al., 2015, Giuntoli et al., 2015a, Giuntoli et al., 2015b). Consequently, two different approaches are followed for the modelling of biogenic GHG emissions. If the mitigation potential of alternative management options were aimed, starting feedstock and re-absorption would be the same for both the reference and the bioenergy system (cancelling out). In this case, all CO_2 emissions are counted, irrespective of their biogenic or fossil origin. If the climate change mitigation for different countries is aimed, the feedstock changes. In this second case, we have to exclude biogenic-CO_2 emissions from the model. This is equivalent to implicitly assume that either all the biomass in the MSW has an annual growth cycle or it has spent sufficient time in the products pool so that the original biomass plant has fully regrown. Figure 2 gives a summary of all carbon fluxes in the production of biofuels and current MSW refuse management.

Secondly, many studies and current European methodology calculate the climate mitigation potential of biofuels and bioenergy by comparing the supply chain impact of biofuels production with the supply chain impact of a fossil comparator. This is not appropriate since, besides the fossil comparator, the reference system should also include the current uses of the feedstock (in this case MSW refuse).

Finally, since biomass decomposition in the landfill is a dynamic process (EUR 27215 EN, 2015), a dynamic LCA assessment is also required. This is similar to what has been done for forest residues degradation (Giuntoli et al., 2015b, IPCC, 2013, Pingoud et al., 2012, Sathre and Gustavsson, 2012). The standard, normalised characterisation factors applied in LCA analysis, Global Warming Potential (GWP 100), are not appropriate to capture these transient phenomena (Giuntoli et al., 2015b). Therefore, firstly we apply the absolute formulation of the climate metric, Absolute Global Warming Potential (AGWP) to capture the dynamic trends. Secondly, we use two different types of metric: i) the AGWP is a cumulative metric that relates to certain climate change impacts such as sea level rise; ii) the Absolute Global surface Temperature change Potential (AGTP), which is an instantaneous metric and more appropriate to represent climate impacts associated to the temperature anomaly, such as extreme weather events.

In this study, only Well Mixed GHG (WMGHG) including CO_2, CH_4 and N_2O are considered. However, Near Term Climate Forcers (NTCF) such as aerosols and ozone precursors may have an important influence on climate, although this often results in a net cooling. Since most of the NTCFs are also local, air pollutants and future strategies will likely limit their emissions. Consequently, by excluding their impact we are applying a conservative assumption for the production of biofuels from MSW refuse. Construction and dismantling of the thermochemical biorefinery and the ashes management from biofuel production and incineration are not included in the assessment and they are assumed to contribute equally for all regions in Europe. The modeling and calculation of the results is done using spreadsheets.

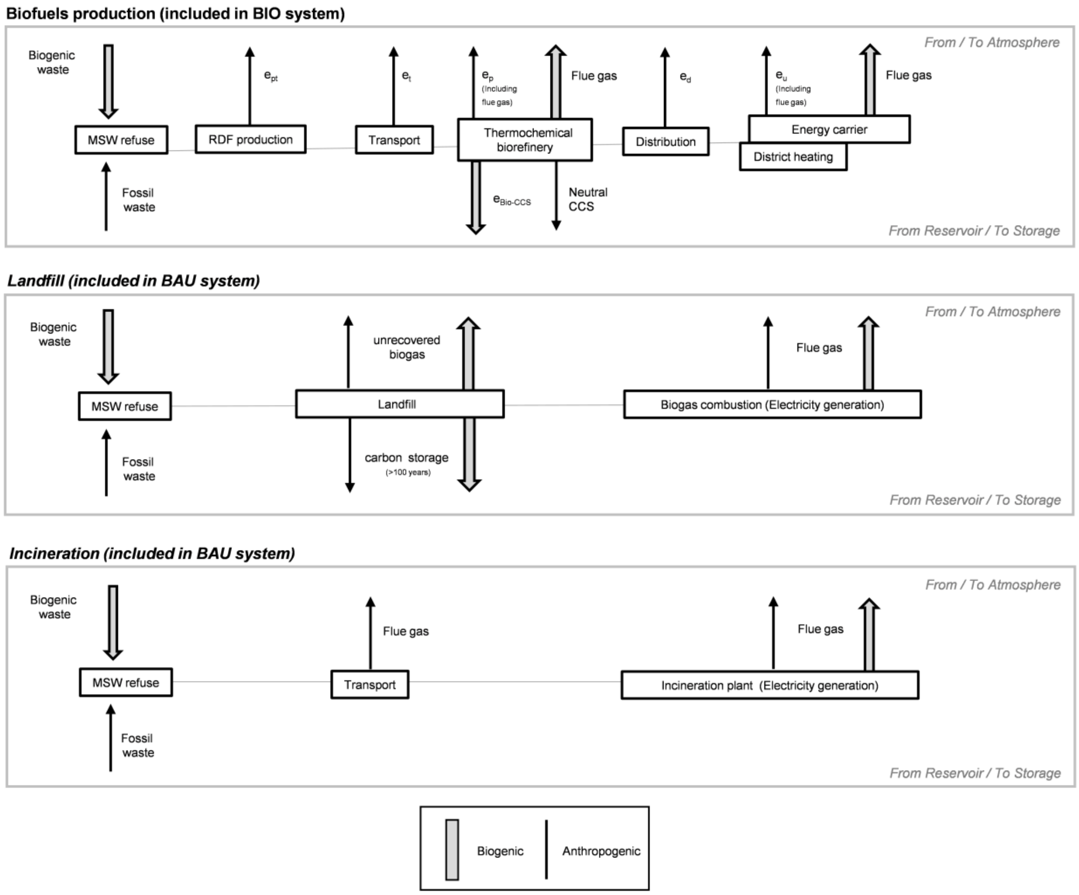

Figure 2. Carbon fluxes associated to the production of biofuels from MSW refuse (included in the bioenergy, BIO, system), the landfill disposal and the incineration with energy recovery (included in business as usual, BAU, system).

3.1. Definition of the bioenergy (BIO) and business as usual (BAU) systems

Two systems are defined in this study: the bioenergy system (BIO) and the business as usual system (BAU). Both systems are based on the same amount of MSW refuse and an equal amount of all products is generated (Figure 3). The business as usual system (BAU) includes the current management of MSW refuse through landfilling and/or incineration and the production of transportation fuels from fossil fuels. Both landfill and incineration options include energy recovery (via biogas combustion in an engine and a boiler respectively). However, only incineration provides heat for district heating if necessary (common practice in Northern Europe). In the bioenergy system, the MSW refuse is used to produce biofuels and electricity, and district heating if necessary (using waste heat from the biorefinery). Since there is a deficit of electricity compared to the amount produced in the BAU system, this has to be balanced from the electricity grid.

In the modelling of the BAU system, a fraction of the generated biogas leaves out contributing to climate change and the rest is used as fuel to generate electricity or burned in a flare. The guidelines for the assessment of the timing of GHG emissions have been provided by the IPCC (IPCC, 2013). For incineration, however, the emissions are evenly produced, as well as in the thermochemical biorefinery. Further details are given in the SM.

3.2. Geographical scope of the assessment

In order to help decision-makers to analyse the environmental risks and benefits of biofuels production from MSW refuse, two extreme examples of current MSW management systems in Europe are assessed: Spain, where landfilling is dominant and incineration is below European average levels; and Sweden, where landfilling is negligible and incineration is dominant (Figure 4). In both Spanish and Swedish cases, the incorporation of Bio-CCS is assessed.

Table 1 shows the typical composition of the MSW refuse in Spain and Sweden. It can be seen that the content of biogenic carbon depends on the waste composition and the collection system, having Spanish MSW refuse a higher biogenic fraction than Sweden. Therefore, the biogenic content is, together with MSW management systems, an important parameter in the assessment.

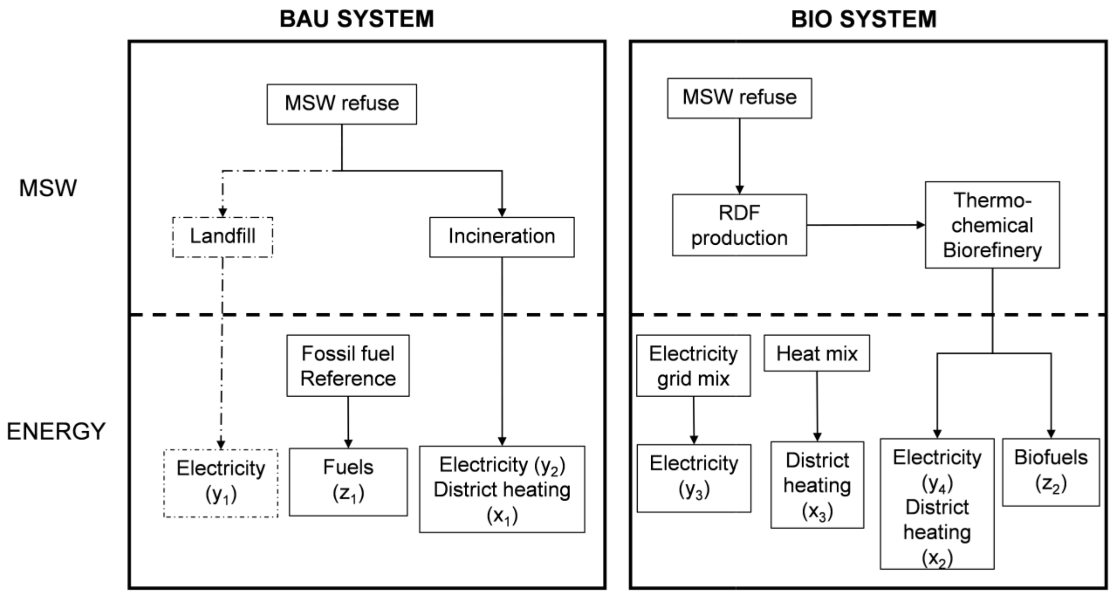

Figure 3. Definition of the bioenergy and business as usual systems in the study. The same mix of fuels (z_1=z_2), electricity (y_1+y_2=y_3+y_4) and district heating (x_1=x_2) are produced in both systems. The x_i, y_i and z_i values represent the shares in LHV basis.

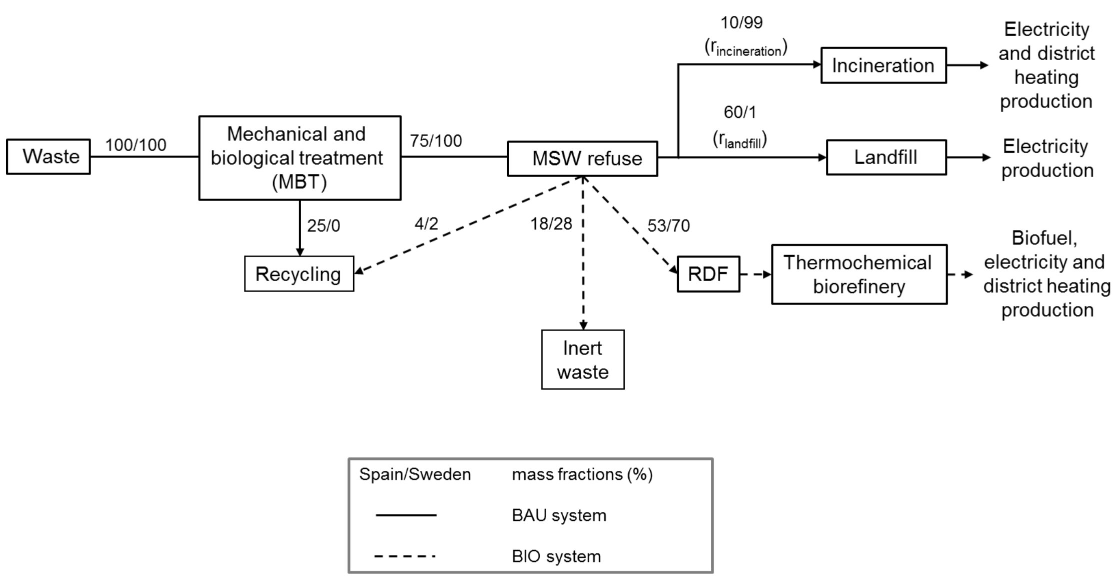

Figure 4. Typical management of unsorted waste in Europe along with the proposed production of biofuels in this study. The given values (mass basis “as received”) represent the Spanish and Swedish management systems respectively.

Table 1. Composition of MSW refuse in Spain and Sweden in % mass basis. The values in brackets are expressed in % carbon basis (Jones et al., 2013, IEA, 2012).

	Total carbon	Biogenic fraction	MSW refuse composition		Biogenic carbon	
			Spain	Sweden	Spain	Sweden
Organic matter	48	(100)	49	31	20	15
Paper-cardboard	44	(99)	19	23	6	10
Plastics	60	(0)	12	14	0	-
Textiles	55	(50)	4	12	2	3
Wood	50	(100)	1	-	1	
Hygiene products	50	(36)	-	12	-	2
Inert waste	3	(0)	16	8	0	-
Total					**29 (76)**	**31 (66)**

Table 2. Energy and material balance of the thermochemical biorefinery producing biofuels from RDF.

<table>
<tr><th colspan="2">Input[a]</th><th colspan="2">Process</th><th colspan="2">Output</th></tr>
<tr><td>RDF (MW_{th})</td><td>100</td><td>Net efficiency</td><td>35% [b]</td><td>Ethanol (Mt/yr)</td><td>23.7</td></tr>
<tr><td>RDF (t/h)</td><td>22.5</td><td>Electricity consumption in RDF production (GWh/yr)</td><td>8.2</td><td>DME (Mt/yr)</td><td>6.6</td></tr>
<tr><td>LHV (MJ/kg)</td><td>16</td><td rowspan="2">Waste heat available for district heating (GWh/yr)</td><td rowspan="2">105.5</td><td>Net electricity (GWh/yr)</td><td>34.8</td></tr>
<tr><td>TOTAL (TJ/yr)</td><td>2843</td><td>TOTAL (TJ/yr)</td><td>995</td></tr>
<tr><th colspan="6">Incorporation of Bio-CCS</th></tr>
<tr><th colspan="4">Process</th><th colspan="2">Output</th></tr>
<tr><td colspan="3">Electricity consumption in the Bio-CCS process (GWh/yr)</td><td>4.6</td><td></td><td></td></tr>
<tr><td colspan="3">Carbon captured in the Bio-CCS process (t carbon/h)</td><td>1.5</td><td>Net electricity (GWh/yr)</td><td>30.2</td></tr>
<tr><td colspan="3">Carbon captured relative to emitted in the thermochemical biorefinery</td><td>25%</td><td>TOTAL (TJ/yr)</td><td>978</td></tr>
</table>

[a] The efficiency in the conversion of MSW refuse in RDF is assumed to be 70% (Arena et al., 2015, Yassin et al., 2009).

[b] The net efficiency increases up to 40% if district heating is produced.

3.3. Modelling of the thermochemical biorefinery

For the life cycle inventory of the biorefinery, we rely on a previous work where we modelled a thermochemical biorefinery producing dimethyl ether (DME) and ethanol from lignocellulosic biomass (Haro et al., 2013a and 2013b). In the same work, we explored the potential of CO_2 capture and storage (i.e., Bio-CCS). For the production of biofuels, the MSW refuse has to be pre-treated and converted into a solid fuel, called either refuse derived fuel (RDF) or solid recovered fuels (SRF), which is then further processed in a thermochemical biorefinery. In this study, the term RDF is preferred to SRF, since the latter applies to a European Standard for the use of the fuel in conventional energy applications, e.g. co-firing in power plants (EN 15357:2012). The pre-treatment of MSW refuse consists of a shredder to reduce the particle size, a trommel to separate small particles, and a magnetic separator and an eddy current separator for ferrous and non-ferrous metals recovery. Finally, the RDF is pelletised to increase the density in order to allow a better handling (Caputo and Pelagagge, 2002a and 2002b). RDF from MSW refuse usually has a heating value of about 16 MJ/kg (Arena et al., 2015, Onel et al., 2014, Yassin et al., 2009). Table 2 shows the energy and material balance of the biorefinery. Electricity surplus is available as a co-product in the process. The difference between the thermochemical biorefinery with and without Bio-CCS incorporation is in the total net electricity production since some of the produced electricity is consumed in the conditioning of CO_2 capture (Boot-Handford et al., 2014, Von Der Assen et al., 2013).

3.4. Static LCA assessment: GHG balance and differential GHG impact

3.4.1. GHG balance and saving

The GHG balance ($E_{biofuel}$) is defined as an annual average of all anthropogenic cradle-to-grave GHG emissions in the production of biofuels using MSW refuse. The biogenic carbon stored in landfill and from Bio-CCS incorporation is modelled as a negative contribution. The methodology for the calculation has been previously discussed by the authors using lignocellulosic biomass (Haro et al., 2015), using the standard LCA characterization method, GWP(100), and characterisation factors defined by IPCC AR5 (see part 2 in SM). However, when using MSW refuse, it needs to be extended to include the fossil fraction in MSW refuse (IEA, 2012, Jones et al., 2013) (see part 3.1 in SM). The saving of GHG in the production of biofuel (compared to emissions from transportation fuels) is calculated using the guidelines from RED (see part 3.1 in SM).

3.4.2. Differential GHG impact

The differential GHG impact (Equation 1) compares, in terms of GHG emissions reduction, the use of MSW refuse for the production of biofuels with the reference system (BAU) (Gaudreault and Miner, 2015). It differs from the GHG balance in the consideration of the displaced and/or avoided emissions due to both material and energy substitution (i.e., it includes the burdens). The GHG impact of the BIO (E_{BIO}) and BAU (E_{BAU}) systems are calculated using Equations 2 and 3 respectively. Moreover, the standard LCA characterisation method, GWP(100), and characterisation factors defined by IPCC AR5 (see part 3.2. and 3.3. in SM) are also used. Avoided emissions are represented by a fossil reference value (EF) according to the mix of products from the biorefinery. The EF value for fossil transportation fuels is 90.3 g CO_2/MJ according to the latest recommendation from the Joint Research Centre (JRC) (Directive 2015/1513). For electricity, the fossil reference is the average GHG emissions of the grid mix of the assessed country (CO_2 Score card, EUR 27215 EN, Spanish Electric Net) (Table 3). The EF value for electricity is used in the bioenergy system to calculate the fossil emissions from the electricity grid to balance the electricity production in the BAU system (y_3). The EF value for transportation fuels allows calculating the GHG emissions from the production of fossil transportation fuels (z_1).

$$\text{Differential GHG impact} = E_{BIO} - E_{BAU} \quad (1)$$

Table 3. Fossil references for fuel and electricity production and shares of electricity and biofuel production (LHV basis) in BIO and BAU systems from Figure 3.

	Spain	**Sweden**
EF_{fuel} (Directive 2015/1513)(g CO_2 eq./MJ)	90.3	
$EF_{electricity}$ (EUR 27215 EN, 2015) (g CO_2 eq./MJ)	110.7	17.5
EF_{heat} (System perpectives on biorefineries, 2014) (g CO_2 eq./MJ)	-	-68
x_1	0	52%
x_2	0	9%
x_3	0	43%
y_1	10%	0%
y_2	37%	47%
y_3	38%	15%
y_4	9%	5%
$z_{1,2}$	53%	28%

$$E_{BIO}=E_{biofuel}\cdot(z_2+y_4+x_2)+EF_{electricity}\cdot y_3+EF_{heat}\cdot x_3 \quad (2)$$

$$E_{BAU}=EF_{fuel}\cdot z_1+(E_{landfill}\cdot r_{landfill}+E_{incineration}\cdot r_{incineration})\cdot(x_1+y_1+y_2) \quad (3)$$

$E_{landfill}$ represents the GHG emissions from the landfill, which depend on the behaviour of landfilled materials. Figure S1 in the SM shows the parameters assumed in this study. Although IPCC and EPA establish default values, some parameters are country-specific and even vary between regions (see part 3.2 in SM) (EPA, 2014, EUR 24708 EN, 2010, IPCC, 2006). $E_{incineration}$ represents the GHG emissions from MSW refuse incineration (see part 3.3 in SM). The results in terms of grams of CO_2 equivalent per ton of MSW refuse are corrected by the system efficiency (Equation 4) to use the same functional unit in both systems: 1 MJ of total products from the biorefinery. The purpose of this correction factor is to include the impact of the different conversion efficiencies in the BAU system compared with the BIO system.

$$E_i = \frac{\text{g } CO_2 \text{ eq}}{\text{t MSW refuse}} \cdot \frac{\text{t MSW refuse/year}}{(\text{MJoutput/year}) \cdot \left(\frac{\eta_i}{\eta \text{ biorefinery}}\right)} \tag{4}$$

where i is landfill or incineration

3.5. Dynamic assessment: CMI and DCI

To calculate the time-dependent climate mitigation potential, each individual WMGHG has been modelled in BIO and BAU systems. The calculation is based on two climate metrics: AGWP and AGTP. IPCC methodology is used and parameters for each individual WMGHG are taken from IPCC AR5.

3.5.1. CMI

The climate mitigation index (CMI) assesses the mitigation potential of producing biofuels from MSW refuse instead of following current MSW management systems. The CMI is dimensionless. For each region assessed (Spain or Sweden), the feedstock is identical independently of how the MSW refuse is disposed of. Therefore, since the re-absorption factor is equal in both systems, all GHG emissions (CO_2 included) are counted from the MSW refuse for both biogenic and fossil origin. The CMI is calculated according to Equation 5 (Pingoud et al., 2012, IEA 2012). The values of AGWP for BIO and BAU systems are calculated from the annual emissions of each WMGHG as explained elsewhere (Giuntoli et al., 2015b, IPCC, 2013, Sathre and Gustavsson, 2012) (see part 4 in SM).

Regarding the values of CMI, it is possible to compare the behaviour of the BIO system with the current MSW management, and electricity and transportation fuels (i.e., the BAU system) for a specific region (Scheme 1). Since the CMI is based on the AGWP values, this comparison gives the cumulative climate mitigation of producing biofuels. Therefore, if the CMI reaches a positive value at a certain time, it means that at this time there is no accumulated climate benefit, i.e., the BIO system starts to be worse than the BAU system. For negative CMI values, there is an accumulated climate benefit, until the CMI reaches -1 when then the BIO system has no emissions of WMGHG.

$$CMI=\frac{(AGWP_{BIO}-AGWP_{BAU})}{AGWP_{BAU}} \quad (5)$$

$$CMI>0 \rightarrow AGWP_{BIO}>AGWP_{BAU} \rightarrow \text{Climate worsening}$$

$$CMI=0 \rightarrow AGWP_{BIO}=AGWP_{BAU} \rightarrow \text{Climate neutral}$$

$$-1<CMI<0 \rightarrow AGWP_{BIO}<AGWP_{BAU} \rightarrow \text{Climate mitigation}$$

$$CMI=-1 \rightarrow AGWP_{BIO}=0 \rightarrow \text{BIO is climate neutral}$$

(**Scheme 1**)

3.5.2. DCI

The differential climate impact (DCI) measures the climate benefit in the production of biofuels from MSW refuse in order to compare the results of regions with different waste management systems. The units of DCI are K·kg MSW refuse^{-1}. In different regions, feedstock composition differs and therefore the biogenic emissions cannot be modelled as for the CMI (Giuntoli et al., 2015a). As mentioned above, in this case it is only possible to account for fossil carbon and biogenic non-CO_2 emissions. In the same way, the biogenic carbon stored in landfill and from Bio-CCS incorporation are modelled as negative contributions. The DCI is based on the AGTP metric as the surface temperature response to the replacement of BAU by BIO (Equation 6). The values of AGTP for BIO and BAU systems are calculated from the annual emissions of each WMGHG as explained elsewhere (Giuntoli et al., 2015b, IPCC, 2013, Sathre and Gustavsson, 2012) (see part 4 in SM).

Regarding the values of the DCI, a direct comparison of two different regions can be made. Since the DCI is based on AGTP values, the comparison gives the climate benefit at a specific time; an instantaneous comparison that is not influenced by the accumulated effect of previous WMGHG emissions. Therefore, if the DCI of one region is lower than in another region, it means that there is a larger climate benefit in the production of biofuels in this region at a specific time.

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

3.6. Scenarios modelled

Two scenarios for current MSW refuse management, transportation fuels and power sector are considered (i.e., BAU system). In both scenarios, the production of biofuels in the BIO system is considered to be continuous; that is 1 MJ of products (biofuels and electricity) is produced every year.

For the two BAU systems, we consider two hypothetical future evolutions:

- *Scenario 1*. The production of biofuels is continuous. In this scenario, we assume that the BAU system, i.e., MSW management system and energy mix (transportation fuels and electricity) do not change for the whole period. This scenario applies for both the static and dynamic assessments. For further details, see part 3 in SM.
- *Scenario 2*. It considers an evolution of the BAU system according to the legal targets and recommendations set by the European Commission. This scenario applies only for the time-dependent assessment since it involves an evolution of the BAU system. This evolution brings a landfill-banned BAU system and would also be closer to the future evolution of MSW management in Europe (ETP, 2010, ETP, 2012, Slade et al., 2011 WEO, 2014). Considering the selected regions, the targets set in the landfill and the waste framework Directives have been already achieved in Sweden but not in Spain (Council Directive 1999/31/EC, Directive 2008/98/EC). Therefore, Spain should reduce the amount of biodegradable municipal waste in MSW refuse and its landfilling rate (Council Directive 1999/31/EC). The Spanish National Framework Plan for Waste Management 2015-2020 establishes the baselines for the future MSW management in an attempt to meet the European targets (State Framework Plan 2015-2020). We assume a delay of 5 years in fulfilling these requirements (Figure 5 and part 5 in SM). Therefore, the targets set in the national plan are used to define the evolution of the MSW management system in Spain for the first 5 years, e.g. 50% of recycling, 35% of landfilling and 15% of energy recovery. From year 2025 to 2120, we propose 1% of landfilling, 65% of recycling and 34% of energy recovery. As Sweden is closer to the European targets than Spain, we consider an objective of 1% of landfilling, 65% of recycling and 34% of energy recovery in the year 2120. In both countries, the increase

of recycling rates is expected with the introduction of new technologies improving efficiency and sorting capacity. Therefore, we assumed the same MSW management system for both countries in the long term.

Although only a time-dependent assessment can include the impact of scenario 2, it is necessary to adapt the data used in a conventional stationary assessment to get the annual emissions of each WMGHG. In Figure 6, it can be seen that landfilling emissions decrease progressively due to the increase of incineration and recycling rates. Because of this, incineration and biofuel production emissions rise in Spain, whereas they keep practically stable in Sweden. In relation to the energy mix, in Sweden the average CO_2 emissions per MWh of electricity has been practically constant in the last decades (0.063 t fossil CO_2/MWh_e) (EUR 27215 EN). This value is much lower than the emissions in Spain (0.398 t fossil CO_2/MWh_e) (EUR 27215 EN) because of the larger share of nuclear and renewable energy in Sweden. Therefore, we assumed Sweden would keep constant emissions and Spain would gradually reduce its emissions until both countries reach the same level in 2120. Likewise, GHG emissions from district heating in Sweden would also keep constant.

4. Results

4.1. Static assessment

4.1.1. GHG balance and saving

Table 4 shows the results of GHG balance ($E_{biofuel}$) and saving achieved according to the current European regulation. The individual contribution of each parameter to the results is shown in Table S13 and Figure S3 in SM. The GHG balance is lower for Spain due to the lower carbon content and the higher biogenic content in MSW refuse, achieving a saving of GHG emissions compared with the fossil reference. If Bio-CCS was incorporated, the saving would be above the target for biofuels in 2018 (60%) (Directive 2015/1513). For Sweden, there would not be any saving even if Bio-CCS was incorporated.

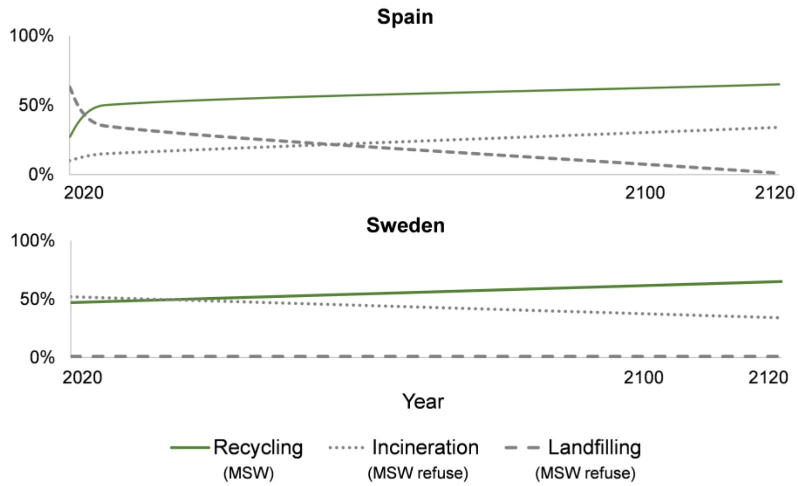

Figure 5. Forecast for the Spanish and Swedish MSW management systems in scenario 2. Data are expressed in terms of the main WMGHG released.

Table 4. Results of the GHG balance and saving in the production of biofuels, calculated according to EU regulation.

Parameters	**Without Bio-CCS**		**With Bio-CCS**	
	Spain	**Sweden**	**Spain**	**Sweden**
$E_{biofuel}$ (g CO_2 eq./MJ)	69	110	28	71
Saving (%)	4	-257	61	-130

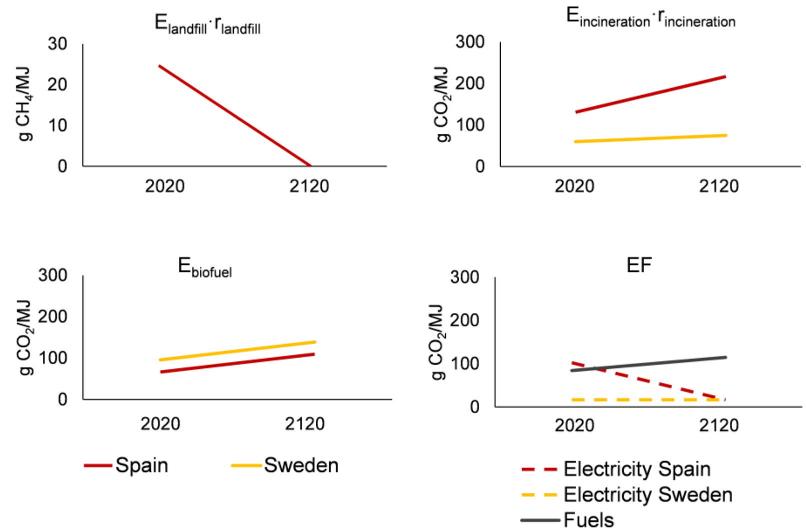

Figure 6. Evolution of emissions from the BAU system in scenario 2. For clarity, the yearly emissions of each WMGHG are combined and expressed in g CO_2 eq./MJ. For a detailed evolution of each WMGHG, see Tables S11 and S12 in SM.

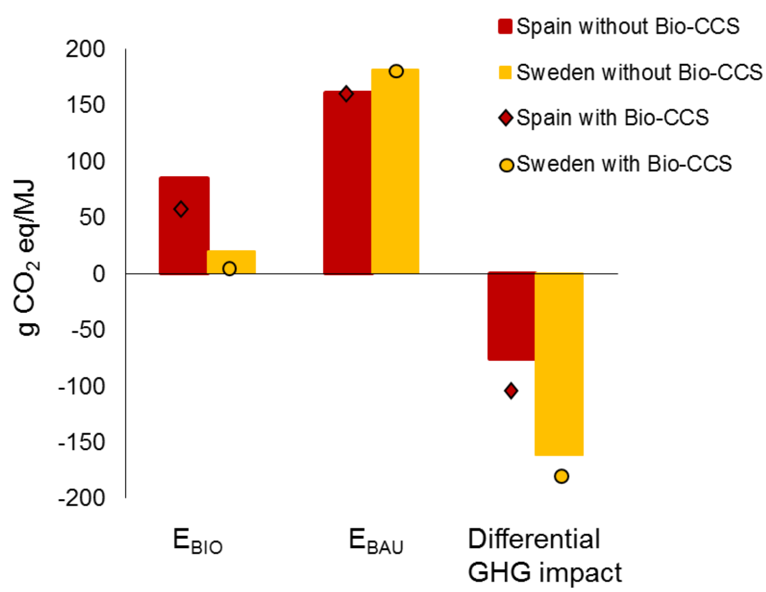

Figure 7. Results of the differential GHG impact in the production of biofuel from MSW refuse (scenario 1).

4.1.2. Differential GHG impact

Figure 7 shows the results for the differential GHG impact, which includes the comparison with the BAU system. It can be seen that the GHG impact of the bioenergy system is higher in Spain than in Sweden since the lower emissions in the production of the biofuel in Spain (where there is a higher biogenic fraction in the MSW refuse) cannot balance the lower emission factor for the Swedish electricity grid and the negative emission factor for the Swedish heat mix (see Table 3). The GHG impact of the BAU system is higher for Sweden because of the higher fossil fraction of the MSW refuse. The differential GHG impact is negative for both countries (i.e., a positive climate impact) but higher in the case of Sweden (-161 g CO_2 eq./MJ) than in Spain (-76 g CO_2 eq./MJ) since district heating producing results in a clear advantage from the point of view of GHG reduction. In both cases, Bio-CCS incorporation involves a similar reduction (around 40 g CO_2 eq./MJ) of the climate impact.

4.2. Time-dependent assessment: CMI

4.2.1. Scenario 1

Figure 8 shows the CMI values for scenario 1. In the case of Spain, there is a sharp reduction of the index from positive to negative and subsequent stabilisation. There is no mitigation until 5 years after the beginning of biofuels production, when up to 45% mitigation is achieved in 2040. Since the transient emissions from the landfill are concentrated around 20 years after the landfilling of the MSW refuse (*landfill memory*, see part 3 in SM), this behaviour was already expected. The mitigation is then reduced until 2120, when the CMI would be slightly positive (4% worse than the BAU system). Therefore, there is no long-term climate benefit in the production of biofuels from MSW refuse in Spain. Considering Sweden, the climate change mitigation is obtained for the whole period considered, where an almost constantly mitigation of 41% is achieved. The mitigation for Sweden is higher than the Spanish one in the first 12 years and after year 2050. Therefore, the production of biofuels would only achieve a larger climate mitigation compared with the BAU system in Spain in the medium term.

Comparing these results with the static assessment, it is clear that these results could not have been predicted from the GHG saving or differential impact. For instance, the differential GHG impact gave a higher climate benefit for Sweden. The static assessment gives an underestimation of the climate benefit in the production of biofuels in a landfill-dominant region like Spain. The results of incorporating Bio-CCS are also different from the stationary assessment. The effect of Bio-CCS incorporation is slightly lower in the time-dependent assessment.

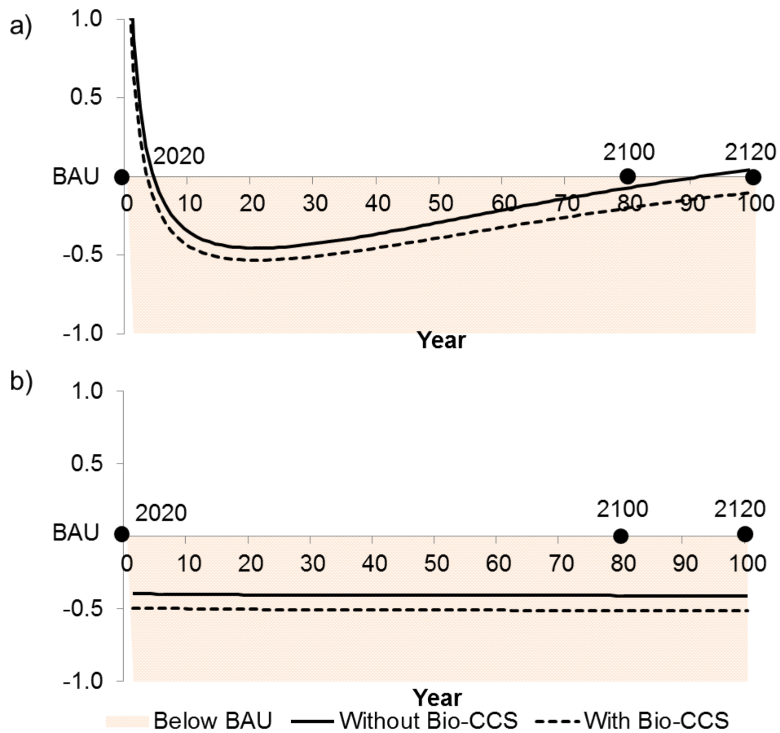

Figure 8. Climate mitigation index for Spain (a) and Sweden (b) with and without Bio-CCS incorporation in scenario 1 where y-axis is CMI, x-axis is year. For clarity, the year 0 is not represented.

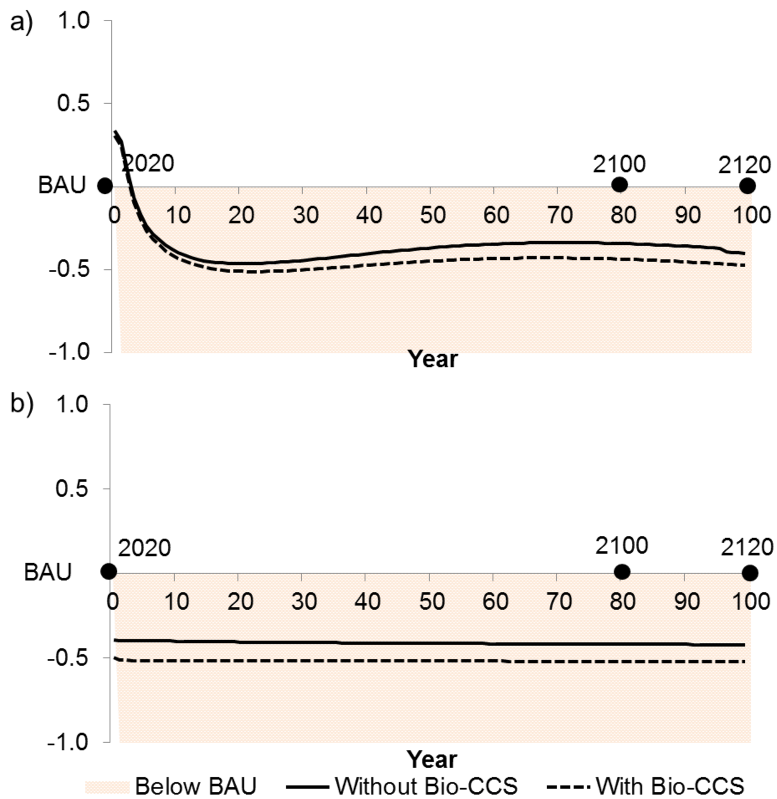

Figure 9. Climate mitigation index in Spain (a) and Sweden (b) with and without Bio-CCS incorporation for scenario 2 where y-axis is CMI, x-axis is year. For clarity, the year 0 is not represented.

4.2.2. Scenario 2

Figure 9 shows the CMI values for scenario 2. For Sweden, the differences between scenario 1 and 2 are minimal. This was expected since Sweden is already close to achieve the proposed targets for MSW management and electricity production, so only the evolution in the emissions from fossil transportation fuels is affecting the results (with a minor impact). However, for Spain, an important difference can be seen in the trend from the beginning because of this evolution of the BAU system. Compared with scenario 1, Spain faces a landfill banning that has a larger impact on the results. Since the landfilling rate is drastically reduced at an early stage, the sharp decrease of the index values is less than in scenario 1 (over half). However, the climate mitigation only occurs after 5 years as in scenario 1. The mitigation reaches a maximum of 51% in 2040, slightly above scenario 1. Later, the impact of higher emissions from incineration, which increases its share in

Spanish MSW management, balances the mitigation reduction and in 2090, the mitigation starts to increase again. In 2010, 40% mitigation is achieved for Spain with a positive trend. Not surprisingly, since both countries meet the same targets for the BAU system in this scenario and their climate benefits are equal after the analysed period. The effect of Bio-CCS incorporation is similar to scenario 1.

4.2.3. Sensitivity analysis

There are several parameters affecting the calculation of the CMI, i.e., the fraction of carbon storage when Bio-CCS is incorporated, the efficiency in biofuel production (thermochemical biorefinery) and the efficiency in the collection of biogas from the landfill. For the sake of clarity, only scenario 1 is used in the analysis, although similar trends are expected for scenario 2.

In the selected configuration of thermochemical biorefinery, the available CO_2 for permanent storage is approximately 25% of the total carbon emitted in the plant (Haro et al., 2013a). This amount corresponds to the already captured pure CO_2 from the syngas in the original thermochemical biorefinery (due to process requirements of the biofuel synthesis catalyst) using pre-combustion technologies. However, it is possible to capture almost all CO_2 from the flue gases by increasing both plant complexity and capital and operating costs. Figure 10 shows that the impact of Bio-CCS incorporation is similar in both assessed regions regardless of the MSW management. By increasing the capture rate from 0 to 100% of the total carbon emitted in the thermochemical biorefinery, the climate mitigation also increases greatly. Maximum mitigations of 60 and 97% can be achieved for complete capture in Spain and Sweden respectively in 2120. The incorporation of Bio-CCS at large capture levels would be then a clear option to obtain long-term climate change mitigation irrespective of the MSW management system. However, it would involve the capture of CO_2 from flue gases, using post-combustion technologies, which is currently only considered for large power plants.

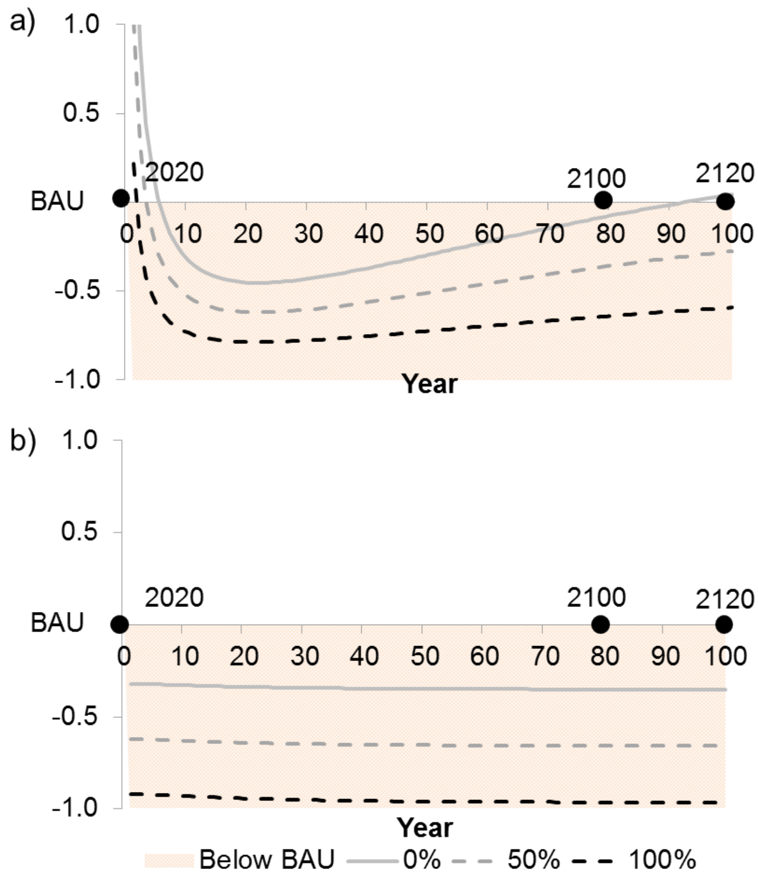

Figure 10. Sensitivity of the climate mitigation index to the carbon captured in the plant for Spain (a) Sweden (b) in scenario 1 where y-axis is CMI, x-axis is year. For clarity, the year 0 is not represented.

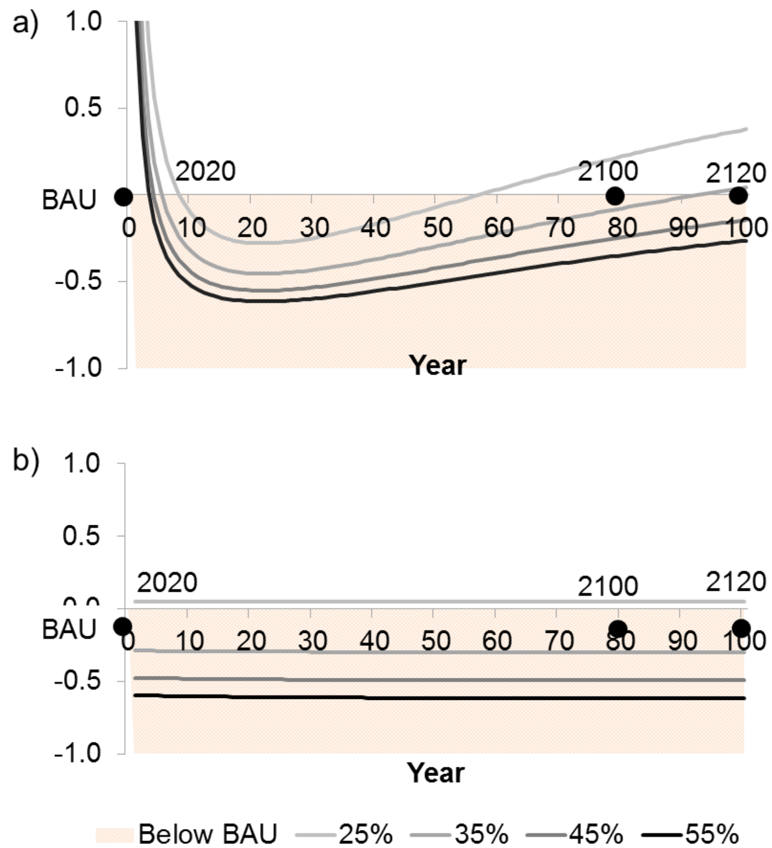

Figure 11. Sensitivity analysis of the mitigation climate index to the energy efficiency of the thermochemical biorefinery for Spain (a) and Sweden (b) in scenario 1 (HHV basis) where y-axis is CMI, x-axis is year. For clarity, the year 0 is not represented.

The efficiency in the production of biofuels (35%, HHV basis, in the selected configuration of thermochemical biorefinery and 40% if district heating is produced) could affect the results. Although it is not the aim of this study to assess technical aspects of thermochemical biorefineries, Figure 11 shows the impact of efficiency on climate mitigation. The impact of the energy efficiency is similar for both countries, as it was expected since the efficiency is proportional to the MSW refuse input to the BIO system (see Eq. 4). Considering the values of efficiency analysed, in the production of liquid biofuels (transportation fuels) typical values range from 35 to 45% (Fernández-Nava et al., 2014). Therefore, the impact of efficiency on the potential climate benefit of producing biofuels from MSW refuse is limited. However, considering the case of Spain, the climate mitigation could be 14% in 2120 (-4% in the base case).

The efficiency in biogas collection in the landfill is the last parameter affecting the CMI results, although only for regions with landfilling, since it is an attribute of the BAU system. A higher collection of biogas in the landfill involves less biogas emissions to the atmosphere. Therefore, it is important to assess the impact of biogas collection (70% in our study). As estimated by the EPA, from 55 to 95% of the biogas produced is collected in modern landfills (EPA, 2014). Figure 12 shows the impact of this variation for Spain, where in case of a very efficient biogas collection (95%), the mitigation potential decreases greatly being zero in 2080 and reaching a worsening of 25% in 2120, which is the worst case in this study for Spain. However, considering specific references for biogas collection in current landfills from Southern and Eastern Europe, no more than 50-55% is actually collected (Aronica et al., 2009, Eriksson and Finnveden, 2009, Fernández-Nava et al., 2014). Considering this, climate mitigation would be 10% in 2120. This value is, however, less than the potential climate benefit considering a reasonable improvement of the energy efficiency in the thermochemical biorefinery, as mentioned above.

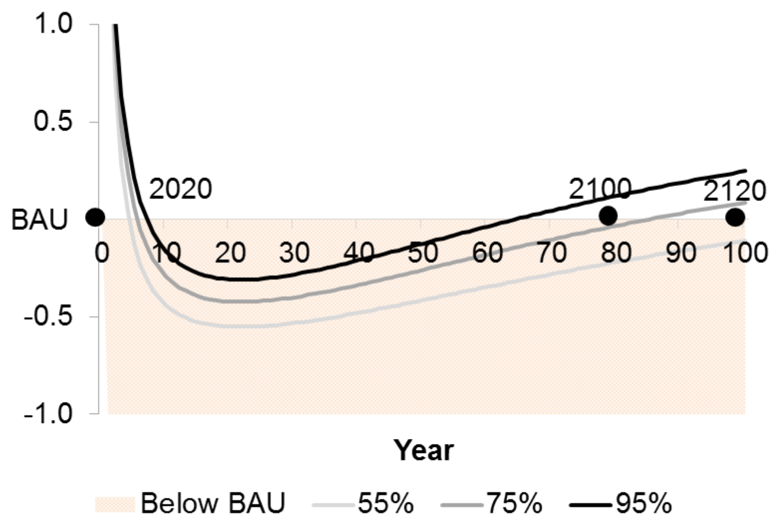

Figure 12. Sensitivity analysis of the climate mitigation index to the biogas collection from the landfill for Spain in scenario 1 where y-axis is CMI, x-axis is year. For clarity, the year 0 is not represented.

4.3. Time-dependent assessment: DCI

4.3.1. Scenario 1

Figure 13a shows the results for the DCI in scenario 1. As opposed to the CMI, the DCI represents an instantaneous metric, so methane impact is rapidly balanced as previously mentioned. The results are in agreement with those of CMI, although some extra information can be obtained here. In the short term, Spain has a potential climate benefit of $-6.5 \cdot 10^{-16}$ K·kg of MSW refuse^{-1} in 2040, whereas Sweden achieves only $-2 \cdot 10^{-16}$ K·kg^{-1}. This makes it even more evident that avoided methane emissions from the landfill only have a short-term impact. Compared with the results for the CMI in scenario 1, the Spanish climate benefit becomes zero in 2080, 30 years before. The reason is the comparison of an instantaneous (DCI) and a cumulative (CMI) metric, where 30 years is the time required to overcome the cumulative climate benefit in the production of biofuels in Spain (the bottom peak). In the long term, biofuel production shows a climate worsening for Spain ($0.6 \cdot 10^{-16}$ K·kg^{-1}), whereas for Sweden it is still $-1.3 \cdot 10^{-16}$ K·kg^{-1}. For instance, only the production of biofuels in Sweden would have a climate benefit in the long term.

4.3.2. Sensitivity analysis

The impact of Bio-CCS incorporation, efficiency in biofuel production and biogas collection efficiency in the calculation of the DCI follow the same trend as in the sensitivity analysis of the CMI (see SM). However, the biogenic fraction in the MSW refuse has an impact on the DCI that cannot be seen in the CMI, since all emissions were equally treated. Therefore, a sensitivity analysis varying the biogenic fraction in MSW refuse from 50 to 100% is presented in Figure 13a. The results are converse depending on the country. For Sweden, the higher the renewable fraction is, the less positive climate impact, since BAU system fossil emissions (incineration) also decrease. Therefore, the use of wastes with high biogenic fraction, e.g. compost, would be discouraged. For Spain, if the renewable fraction increases, so do the landfill emissions, since biogas comes from the biodegradable fraction. The decrease of incineration emissions is not enough to balance the net BAU system emissions (Figure 13b). Therefore, the use of wastes with high biogenic fraction, e.g. compost, would have a positive impact. If the MSW refuse has a large fraction of non-biogenic carbon, e.g. plastics, the permanent storage of this carbon in the landfill balances the climate benefit of producing biofuels.

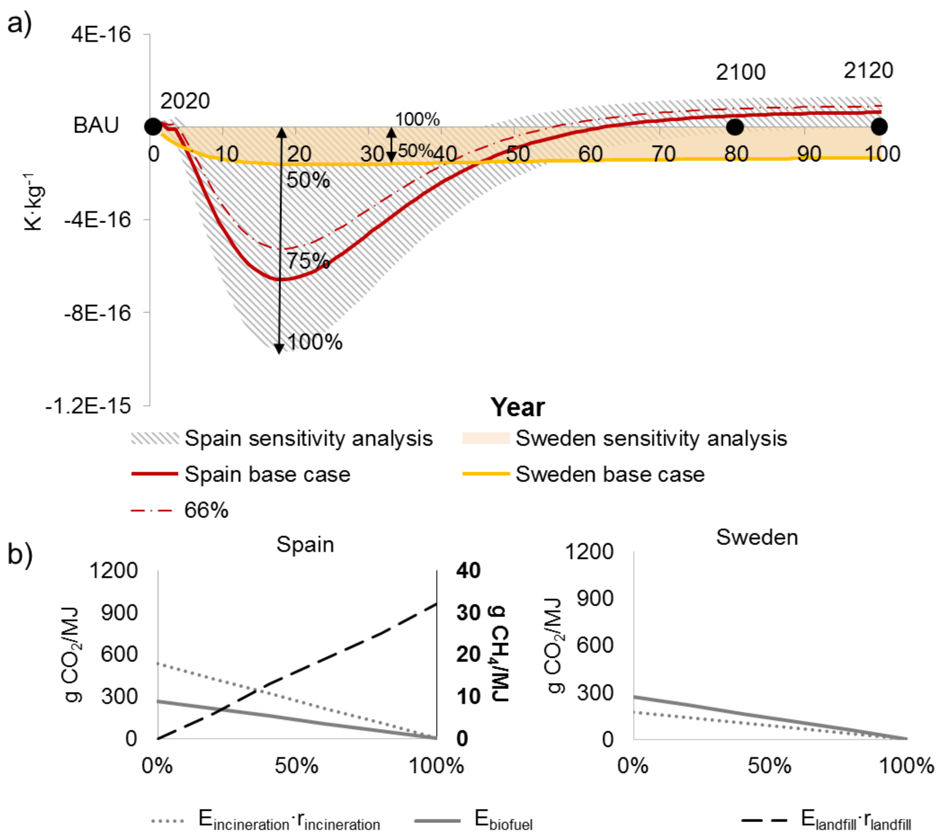

Figure 13. a) Differential climate impact (lines) and sensitivity to the biogenic fraction in MSW refuse (areas) for Spain (grey) and Sweden (brown) in scenario 1 where y-axis is DCI ($K \cdot kg^{-1}$), x-axis is year. The lines represent the base cases and areas of the results of the sensitivity analysis. The dash line represents the Spanish base case if the biogenic fraction was the same than the Swedish. For clarity, the year 0 is not represented. b) Sensitivity of the emissions in BAU and BIO systems to the biogenic fraction (%). The emissions are expressed in terms of the main WMGHG released.

4.3.3. Scenario 2

Figure 14 shows the results for scenario 2 where Spanish BAU system evolves to become the same as the Swedish beyond 2120. The results are in agreement with those of CMI. In Spain, the landfill banning becomes important after 20 years due to landfill disposal decreases until the half of current levels is reached. Moreover, the emissions from the electricity mix are lower than the emissions from incineration, making the total emissions in the BAU system increase. Hence, the maximum climate benefit for Spain is reduced in a

57% compared with scenario 1 in 2037 (2.84 and 6.58·10-16 K·kg-1 respectively), but it is enhanced in the long term since the DCI never becomes positive. At the end of the considered period (2120), the DCI reveals a climate benefit of -1·10-16 K·kg-1. As it happened before, the results for Sweden are the same as in scenario 1.

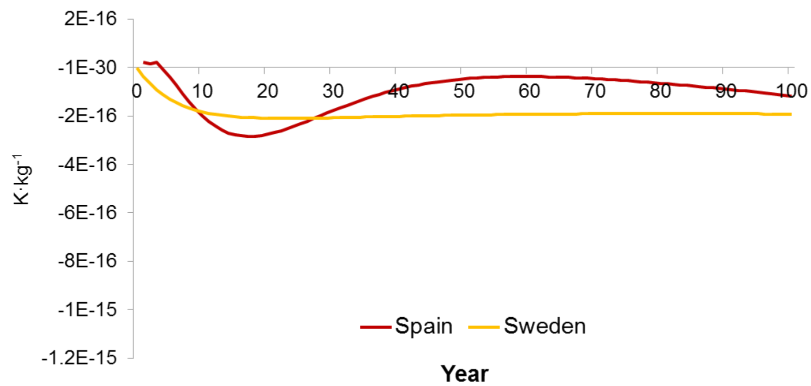

Figure 14. Differential climate impact for Spain (red) and Sweden (yellow) when an evolution of BAU system is considered (scenario 2) where y-axis is DCI ($K{\cdot}kg^{-1}$), x-axis is year.

5. Discussion

This study offers indications of the potential climate change mitigation provided by using MSW refuse as a feedstock for the production of transportation biofuels. In order to provide a complete analysis, both static and time-dependent assessments are illustrated. The static assessment (GHG balance, saving and differential impact) offers clear quantitative results, such as the GHG savings indicator which is currently used in Europe for the certification of biofuels. However, the drawbacks of static life cycle assessments have become apparent in the last years. Static assessments, such as commonly found in the literature, cannot properly capture the impact of landfills in the evolution of GHG emissions with time and thus are unable to properly assess the climate change mitigation of the bioenergy system. Especially when considering potential dynamic evolution of both energy mix and MSW management system in a 100-years period, only the time-dependent assessment can provide a proper impact assessment. This is less evident for the case of a region with current negligible landfilling where the systems considered do not change in time and where the main WMGHG is CO_2 and not a short-lived GHG such as methane.

It is relevant that the climate mitigation achieved by the production of biofuels is similar in the first 30 years for both countries studied, but it diverges in the long term. In regions with current dominant landfilling, in fact, the climate impact after 100 years is equal or slightly worse for the bioenergy system compared to the continuation of the BAU system. However, other relevant impacts and risks associated to the BAU system should be assessed in order to avoid a shift of environmental burdens different from climate change in the production of biofuels from MSW refuse.

We have considered a conservative value for the share of MSW disposal in modern landfills (70%) in Spain (Chacartegui et al., 2015) in order to compare processes at a similar readiness level. The production of biofuels would result even more advantageous if the substitution of the landfills with the worst biogas collection efficiencies in Europe was prioritised. For the incineration, conservative efficiency values for new incineration plants with energy recovery have also been used. The capture efficiency for Bio-CCS incorporation in this study is 25%; this value corresponds to the amount of pure CO_2 that needs to be captured in the biorefinery to guarantee proper functioning of the catalytic synthesis downstream (Haro et al., 2013a). This capture efficiency is above typical values in biochemical production of ethanol (11-13%) (Bio-CCS, IEA-GHG, 2011). A higher capture efficiency would require an increase of both investment and operational costs to capture the carbon present in flue gases (i.e., post-combustion technologies), increasing process complexity and worsening its economy (Haro et al., 2014, Lomax et al., 2015b). Therefore, the impact of Bio-CCS incorporation would be limited in the production of biofuels from MSW refuse. When the impact of the biogenic fraction of MSW refuse was analysed, the results for 100% biogenic fraction are indirectly related with the use of the organic sorted fraction of MSW (usually converted into compost). However, this fraction is currently not promoted for biofuel production (Directive 2008/98/EC).

Finally, some authors consider that the carbon from wood and paper requires a specific treatment for the counting of GHG emissions (Guest et al., 2013 and 2014), and therefore, distinguish them from food waste. However, in this study we consider that this carbon has spent sufficient time in the products pool so that the original biomass plant has fully regrown.

We considered multiple system configurations so to highlight potential improvements and additional risks. This sensitivity analysis shows that the climate mitigation for Spain is uncertain in the long term (ranging from -4 to 14%). Therefore, climate benefit cannot be claimed in the production of biofuels in a current landfill-dominant country, but, and this is crucial, neither a significant climate worsening. The only case providing a clear climate benefit would be in a country with current negligible landfilling coinciding in Europe with countries requiring district heating production, where the climate mitigation could represent 41% (constant for the whole period) compared with the BAU system. However, these results are derived from scenario 1, where the MSW management system of the countries is supposed to keep unchanged for 100 years.

Scenario 2 has analysed an evolution of the BAU system on the assumption of a stable policy strategy for MSW management throughout Europe according to the European targets and in line with the principles of the Circular Economy. The banning of landfilling has revealed as the most significant change. The promotion of landfill banning, despite being a very entrenched practice in Southern and Eastern Europe (Cherubini et al., 2008, Clausen et al., 2016, Fiorentino et al., 2015), would equalise the climate mitigation in the production of biofuels in Spain and Sweden. This result is another example of how the contribution of landfills in climate change is still not fully understood. Instead of being a climate burden because of their methane emissions, it favourably compares with the alternative disposal of MSW refuse (i.e., incineration) because of the storage of biogenic carbon in the landfill. Moreover, the decarbonisation of the transport sector suffers from a lack of alternatives compared with the power sector. The favourable comparison of biofuels production from MSW refuse in a progressively decarbonised BAU system proves that MSW refuse should not be considered as a priority source for electricity production.

6. Conclusions

The production of biofuels from MSW refuse would achieve climate change mitigation in Europe in the medium term. Hence, the substitution of current MSW management (landfill and/or incineration) for the production of biofuels would likely not cause a negative burden for climate change. However, there are important differences for the two extreme examples of current MSW management and electricity pool analysed, Spain (landfilling is dominant, high emissions) and Sweden (landfilling is negligible, low emissions). In Spain, the impact of landfill emissions prevents a climate benefit in the long term, although it does not represent a climate worsening. Therefore, a strategic decision on the MSW management change would mainly rely on avoiding the environmental impacts of landfilling, which are different from those of climate change. In Sweden, the climate benefit is present at all times. Only in the case of landfills with a low biogas collection efficiency, the substitution of the landfill by biofuel production would be clearly positive for the climate in both medium and long terms.

Considering an evolution of the reference system for MSW refuse in Europe (including MSW management, but also electricity and transport sectors), the results become similar for Spain and Sweden in the long term. For instance, in the case of Spain, a clear climate benefit appears. The analysed evolution assumes a progressive banning of landfilling in Europe, decarbonisation of the electricity sector and increase of the emissions from fossil transportation fuels. The favourable comparison of biofuels production from MSW refuse in a progressively decarbonised BAU system proves that MSW refuse should not be considered as a priority source for electricity production but for biofuel production. These results provide policy makers with a scientific basis for the encouragement of MSW refuse in the production of biofuels in Europe.

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Abbreviations

BAU	business as usual (reference system)
BIO	bioenergy
Bio-CCS	carbon capture and storage in bioenergy
DME	dimethyl ether
DOC	degradable organic carbon
EPA	Environmental Protection Agency (USA)
GGR	greenhouse gases removal
GHG	greenhouse gases
IEA	International Energy Agency
IPCC	Intergovernmental Panel on Climate Change
JRC	Joint Research Centre
LCA	Life Cycle Assessment
LHV	Lower Heating Value
MSW	municipal solid waste
RDF	refuse derived fuel
RED	Renewable Energy Directive
SRF	solid recovered fuel
WMGHG	Well-Mixed greenhouse gases

Nomenclature

AGTP	Absolute Global surface Temperature change Potential, $K \cdot kg^{-1}$
AGWP	Absolute Global Warming Potential, $W \cdot year \cdot m^{-2} \cdot kg^{-1}$
CMI	climate mitigation index, —
DCI	differential climate impact, $K \cdot kg$ of MSW refuse^{-1}
EF_i	emissions from the fossil reference for i (transportation fuels or electricity), g CO_2 eq. per MJ
E_i	emissions from MSW refuse in i (landfilling, incineration, BAU system, biofuel production or BIO system), g CO_2 eq. per MJ of product from the biorefinery (biofuel and electricity)
GTP	Global surface Temperature change Potential, —
GWP	Global Warming Potential, —
R	atmospheric decay of a gas, —
RF	Radiative Forcing, $W \cdot m^{-2} \cdot kg^{-1}$
$r_{incineration}$	fraction of MSW refuse incinerated in the region assessed, %
$r_{landfill}$	fraction of MSW refuse landfilled in the region assessed, %
x_i	ratio of heat production (1 BAU system, 2 BIO system)
y_i	ratio of electricity production (1 landfill, 2 incineration in BAU system; 3 electricity mix, 4 biorefinery in BIO system)
z_i	ratio of fuel production (1 BAU system, 2 BIO system)
$\eta_{biorefinery}$	efficiency of the thermochemical biorefinery producing biofuels, % LHV
η_i	efficiency of the electricity generation in landfill or incineration, % LHV

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Paper IV

Contrasting the Greenhouse Gas Removal Potential of CCS and Renewable-Derived Plastics in Advanced Waste-to-Energy Plants

In this paper, the climate mitigation potential of a novel greenhouse gas removal (GGR) technology consisting in the production of renewable-derived plastics from municipal solid waste (MSW) refuse has been evaluated and compared with the option of incorporating Bio-CCS into advanced waste-to-energy (WtE) plants (pre-combustion). It has been submitted for publication to the International Journal of Greenhouse Gas Control. A previous work inspiring this paper was published as conference proceedings in 2016[15].

1. Introduction

Greenhouse gas removal (GGR) technologies are a type of climate engineering aiming to remove greenhouse gases (GHG) from the atmosphere, and thus tackling the root cause of global warming. These techniques either directly remove GHG, typically CO_2, or alternatively seek to influence natural processes to remove GHG indirectly. Integrated Assessment Models (IAMs) are employed to forecast the impact of mitigation technologies on global warming in the next decades (Canadell and Schulze, 2014, Creutzig, 2016, Fuss et al., 2016, Jones et al., 2016, Muratori et al., 2016, Smith et al., 2016, Tokarska and Zickfeld, 2015, Vaughan and Gough, 2016). Several GGR technologies (see Table 1) are being developed aiming to stabilize at 450 ppm CO_2 equivalent concentration (Kriegler et al., 2013). However, IAMs currently mainly focus on Bio-CCS assuming a massive development of this technology in order to achieve the 2 ºC target by 2100 (Kriegler et al., 2013, ETP, 2010).

Bioenergy combined with carbon capture and storage (usually referred as Bio-CCS or BECCS in the literature) can be considered a GGR technology since it allows capturing the biogenic CO_2 produced in industrial processes using biomass or wastes and avoiding its emission to the atmosphere, yielding a net removal of CO_2 from the atmosphere

[15] Aracil C, Giuntoli J, Cristobal J and Haro P.
Proceedings in http://ftp.servcbo.com/1605WASTEENG/Abstracts/Topic7/033-spc0033.pdf

(Koornneef et al., 2012). The IPCC considers this technology to be essential in achieving the 2 ºC target by 2100 (IPCC, 2014). Besides, Bio-CCS has the ability to compensate past and distant emissions and that is why it might be deployed strategically to alleviate the most costly mitigation constraints (Kriegler et al., 2013). However, technical (e.g. the need of a deeper characterization of CO_2 storage sites) and financial limitations are seriously hindering the deployment and implementation of Bio-CCS (Kemper, 2015, ETP, 2015, Lomax et al., 2015, Sigurjonsson et al., 2015), especially at the scales projected in the Representative Concentration Pathways (RCP) scenarios (IPCC, 2013). In line with this, an important concern with Bio-CCS is its dependence on large-scale bioenergy use, which can have adverse impacts on land use and biodiversity (IPCC, 2012). In addition, a debate is ongoing on the excessive reliance of most IAMs in the implementation of Bio-CCS in the short term as the main solution considered to achieve the 2 ºC target by 2100 (Anderson and Peters, 2016a and b, Lackner, 2016). Therefore, it seems appropriate to investigate for alternative GGR technologies able to be implemented in industrial processes using renewable feedstock like biorefineries or advanced waste-to-energy (WtE) plants (plants processing biomass or wastes to produce electricity, heat, fuels and/or chemicals). The production of renewable-derived plastics is proposed in this study as an alternative GGR technology to Bio-CCS. Since biomass cultivation (impacts on land use and biodiversity) might involve uncertainty on the evaluation of the proposed GGR technology, only the use of wastes as feedstock is analyzed.

Renewable-derived plastics, i.e., plastics made from biomass or wastes, represent a biogenic 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 (Sigurjonsson, 2015), renewable-derived plastics can offer a temporary carbon storage thus forming an additional carbon pool (Mclaren, 2012, Shackley and Sohi, 2011). The storage period for a renewable-derived plastic depends not only on the type of plastic, but also on country-specific conditions such as consumer behavior (e.g., lifetime) and the waste management strategy and recycling rates related to the plastic waste (Guest et al., 2013a, Levasseur et al., 2013, Pawelzik et al., 2013, Vogtländer et al., 2014, Wang et al., 2014). Although the carbon storage in renewable-derived plastic materials is not regulated, it can be equated to the carbon storage function of biochar that is already included in the European regulation (Guest et al., 2013a, Von Der Assen et al., 2013, Wang et al., 2014). The storage of biogenic carbon in renewable-derived plastics has been scarcely discussed in the literature (Chen et al., 2016, Haro et al., 2015).

Table 1. List of main available and potential GGR technologies in the literature (Lomax et al., 2015, Smith et al., 2016,). The proposed GGR technology in this study is indicated in bold.

Afforestation and reforestation
Wetland restoration
Agricultural soil sequestration
Biochar
Bio-CCS /BECCS
Direct air capture
Renewable-derived plastics

Renewable-derived plastic materials represent today less than 1% of total plastic consumption in Europe (BIO intelligence service, 2013), but this value is expected to increase at a rate of 20% per year in the next years (Plastics Europe, Plastic waste in the environment, 2011). For instance, the European chemical industry has committed to a gradual increase in the utilization of renewable feedstocks, achieving 25% of renewable-derived chemicals in 2030 (Sustainable biomass in the chemical industry, 2012). This estimation may be reinforced by the fact that the Circular Economy Package adopted by the European Commission foresees, through a revised legislative proposal on waste, the creation of economic incentives for producers to put greener products on the market (e.g. green public procurement and financial instruments such as environmental taxes) (COM(2015) 614 final). However, up to now, most of the renewable-derived chemicals in the market are specialty chemicals (e.g. lactic acid, succinic acid or propanediol based on commodity agricultural products such as sugars and vegetable oils (BIO-TIC project, 2014), but not suitable for massive plastic production (Corma Canos et al., 2007, Fiorentino et al., 2016). There are only a few cases of substitution of fossil plastics by renewable-derived plastics (e.g. starch-based plastic bags, polylactic acid bottles, etc.)

(Dornburg et al., 2004, Piemonte and Gironi, 2011, Yano et al., 2014, Yu and Chen, 2008) and only some attempts at standardization (CEN/TC 411). The reasons for the scarce introduction of renewable-derived plastics are the cost and the security of supply for these new plastic materials. Many technical challenges, especially related to downstream processing, need to be overcome to help promote the use of alternative feedstock streams and reduce processing costs (BIO-TIC project, 2014). Alternatively to the use of new plastic materials, the drop-in approach involves the production of renewable-derived chemicals that can directly substitute the fossil-based chemical equivalent in the production of current plastic materials. Renewable-derived drop-in chemicals can thus be directly introduced into existing and established value chains, infrastructure and markets (Arvidsson, 2016, Haro et al., 2014).

Plastic materials are increasingly widespread in our lives. The use of plastics contributes to societal and economic development reducing transport costs, replacing less safe materials or improving food conservation among others (Plastics Europe, 2015). Nevertheless, the durability, understood as effective use, of plastic materials is variable, depending on the material characteristics and consumers' behavior, e.g. a bottle might be used for a few days while a water supply pipe might be in service for dozens years. The world production of plastics has increased by 30% in the last ten years and Europe is the second largest producer after China. In Europe, 59 million tonnes of plastics were produced and 47.8 million tonnes were consumed in 2014 (Plastics Europe, 2015). Recycling of waste plastics becomes crucial to extend plastics lifetime, reducing the consumption of fossil fuels and the environmental footprint. As an example, in 2010 there were 95.5 billion plastic carrier bags (1.42 Mt) placed on the EU market, most of them (92%) being for single use (COM (2013) 123 final). Depending on the plastic material, there is a maximum number of recycling cycles and therefore a limit to their overall lifetime. However, not all plastic materials are recycled and each recycling cycle generates a waste refuse that cannot be further recycled. Hence, every year around 70% of post-consumer plastics waste is either landfilled or incinerated (with energy recovery) in Europe (Plastics Europe, 2015). These figures do not include the million tonnes of plastic entering the oceans yearly worldwide due to mismanagement of plastic waste disposal (for the EU this value ranges between 0.05 and 0.12 million tonnes per year) (Jambeck et al., 2015). However, plastic pollution on marine ecosystems has detrimental environmental consequences whose extent is still subject of research (Thompson et al., 2009). Therefore, in this study, it is considered that the totality of plastic waste is managed or recycled.

In the evaluation of renewable-derived plastics production as a GGR technology, the impact of the renewable feedstock on climate change needs to be clarified. As mentioned before, the impact of land-use change (both direct and indirect) may have a dramatic impact on the cradle-to-grave GHG emissions of biomass-derived products, as it has also been described for biofuel production (Directive 2015/1513). Wastes are an alternative renewable feedstock to biomass. However, wastes are not always fully biogenic as in the case of industrial and urban wastes. In this study, we consider municipal solid waste (MSW), which is a cheap and widely available source in Europe. MSW is a heterogeneous mix of different biogenic and fossil fractions, which can be separated at source and/or in a mechanical and biological treatment plant. In this study, the fraction evaluated is the MSW refuse, i.e., the unsorted fraction of MSW that cannot be further processed and goes to energy recovery or landfilling (Commission Decision 2000/532/EC). In 2014, 138 million tonnes of MSW refuse were generated in Europe representing the 55% of total MSW production (Eurostat). The MSW refuse generated in Europe has an energy potential similar to agricultural residues (1,250 PJ/year) (Biomass Futures D3.3 report). MSW refuse contains around 10-15% of plastic waste so if MSW refuse was pretreated and used as feedstock to produce more plastics, the loop on plastic recycling would be closed.

2. Goal and scope of this study

The aim of this work is to assess the climate benefit of renewable-derived plastics as a GGR technology and to provide a comparison with CCS incorporation. The selected conversion process is an advanced WtE plant processing MSW refuse. Since MSW refuse is not fully biogenic, i.e., there is a fraction of fossil carbon, the share of fossil carbon in the renewable-derived plastics needs to be properly accounted for. The resulting climate benefit is dynamically assessed considering the waste management schemes in Europe (incineration vs. landfilling) for a period of 100 years and the emissions from the substituted production and disposal of fossil plastics (from crude oil). It is important to remark that renewable-derived plastics (drop-in approach) are disposed at the end of their lifetime following the same path as fossil (conventional) plastics in the waste management scheme. Therefore, the different final disposal techniques of waste plastics have an impact on both the climate impact of the reference case of MSW refuse management and the effective biogenic carbon storage of renewable-derived plastics. This double impact makes it even more important to avoid double counting in the assessment of a GGR technology.

For instance, from a cradle-to-gate point of view, the production of renewable-derived plastics involves a biogenic carbon storage contributing to climate mitigation that does not depend on the timing and technique of its disposal. However, if cradle-to-grave emissions are considered (as they are in this study) the climate mitigation will depend on the final disposal of the plastic waste (timing and technique). In the case of incineration disposal the effective GHG removal of the renewable-derived plastic will depend on its lifetime. In the case of using a partly biogenic feedstock, e.g. MSW reuse, the effective GHG removal is even more dependent on the timing since fossil GHG emissions would be also released during incineration. Therefore, the uncertainties associated with the timing of biogenic carbon storage must be tackled with a time-dependent (dynamic) perspective in order to determine the potential climate change mitigation from renewable-derived plastics.

The production of renewable-derived plastics is an alternative GGR technology never analyzed to date. The idea of rewarding extra-avoided emissions from renewable-derived plastic materials was tentatively assessed in a previous work of the authors (Haro et al., 2015b). In another work (Aracil et al., 2017), the use of MSW refuse in thermochemical biorefineries producing biofuels was shown to have a positive climate impact compared with conventional waste management and disposal in Europe. In this study, the previous works are expanded by analyzing the time-dependent climate impact of renewable-derived plastic production as a GGR technology. In order to give a fair figure of the potential climate impact of renewable-derived plastics, the incorporation of Bio-CCS to the same advanced WtE plants processing MSW refuse is assessed and compared.

Firstly, a comprehensive description of the methods used in the study is provided. A business as usual (BAU) system is defined, where a conventional waste management scheme is described for Europe (Spain and Sweden as case studies) along with the production of transportation fuels and plastics. A bioenergy (BIO) system is described for the production of renewable-derived plastics in an advanced WtE plant (thermochemical biorefinery) using MSW refuse and balancing electricity and heat production from the BAU system. The dynamic modeling of biogenic carbon storage is described accordingly. Secondly, the differential GHG impact of the production of renewable-derived plastics from MSW in an advanced WtE plant is carried out (i.e., integrating the emissions from the different processes involved in both systems) including the option of incorporating Bio-CCS into the plant. Thirdly, the dynamic assessment is carried out including the impact of different waste management schemes and its evolution, biogenic fraction in MSW refuse and Bio-CCS incorporation. Finally, the impact of transportation fuel co-production in the advanced WtE plant is analyzed.

3. Methodology and definition of the systems

3.1. Definition of the Bioenergy (BIO) and Business as Usual (BAU) systems

Figure 1 shows the system boundaries for the BIO and BAU systems analyzed in this study. Both systems are sized to treat the same flow of MSW refuse and produce the same functions (energy products and plastics). 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 (to the grid). It also includes the production of transportation fuels and plastics from fossil fuels. In the BIO system, MSW refuse is used in an advanced WtE plant (a thermochemical biorefinery in this study) to produce renewable-derived plastics (drop-in approach), fuels, electricity and district heating. The configuration of the thermochemical biorefinery is independent of the region assessed. However, since the impacts associated to the production of heat and electricity in the BAU system (y_1, y_2 and x_1) depend on the region assessed, it is necessary to balance the deficit of electricity production from the regional grid and the deficit of heat from the heat mix.

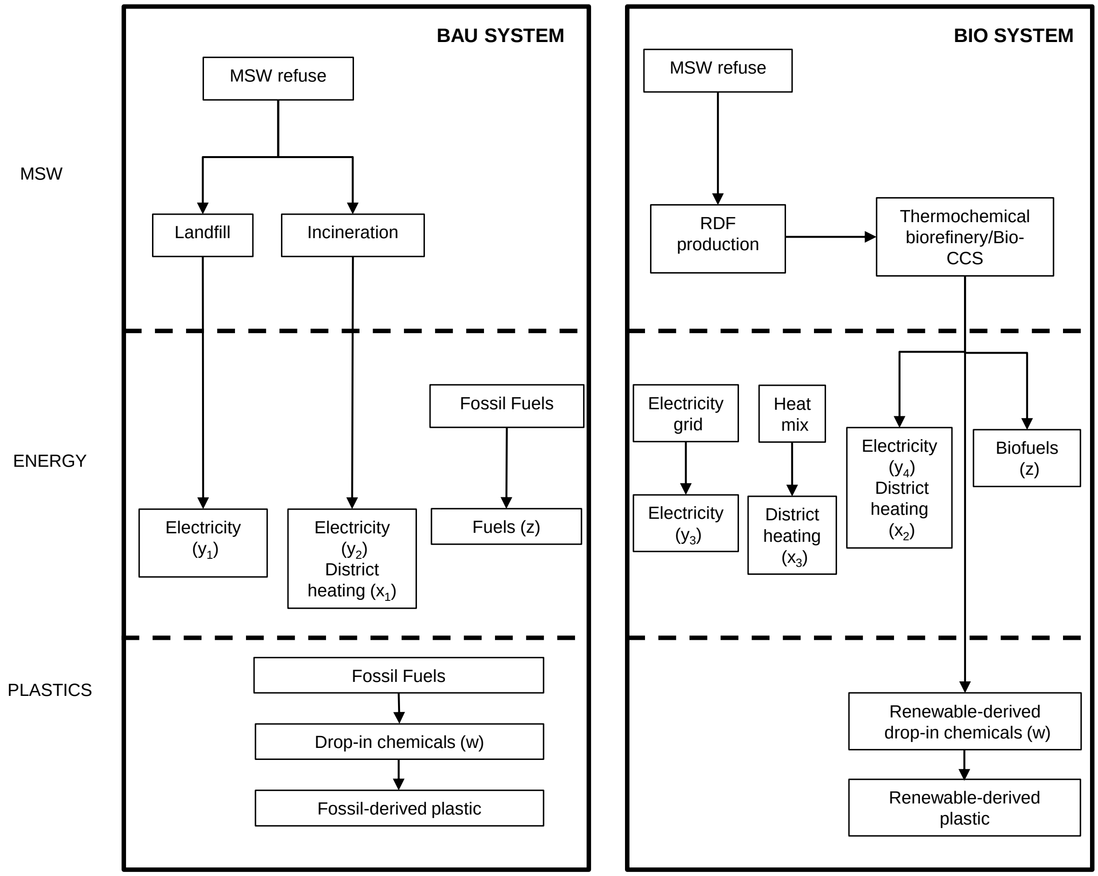

Figure 1. Systems boundaries for the BAU (conventional waste management, and fossil chemicals and transportation fuels production) and the BIO (production of renewable-derived plastics from MSW refuse and balance of electricity and heat, district heating, production from conventional waste management)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

3.2. Modeling of the BAU system

Two European countries (Spain and Sweden) have been selected as representative of the two main waste management schemes in Europe. Sweden represents the case of Northern and Central Europe, where incineration with energy recovery (heat and electricity) is dominant and landfilling is negligible. Spain is representative of the Southern and Eastern European case, where landfilling is dominant and energy recovery is focused on electricity production using landfill gas. Figure 2 shows a comparison of the conventional waste management scheme of the MSW refuse in both countries. In Spain, a higher fraction of MSW refuse is produced since the recycling ratio is lower than in

Sweden (Almasi and Milios, 2013, Milios, 2013, Organic resources and biological treatment, 2010, Vandecasteele et al., 2016).

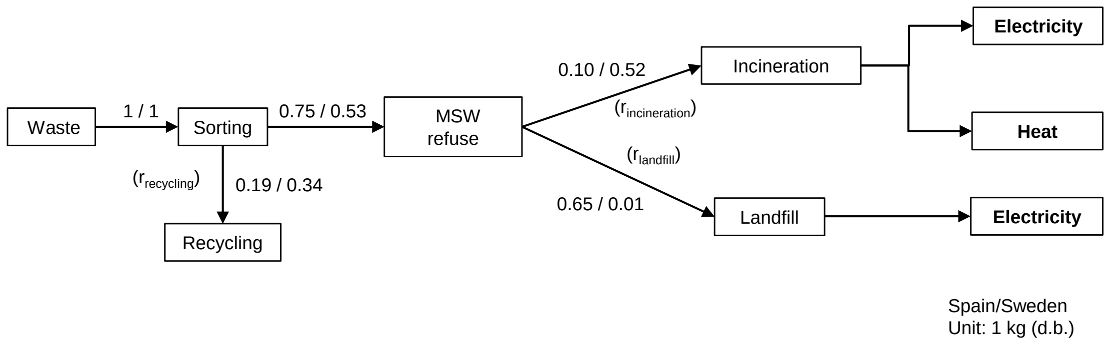

Figure 2. Conventional management of MSW refuse for the selected countries in the study (Spain and Sweden). Final products are shown in bold. Values are in dry mass basis considering 1 kg of MSW refuse as input (Almasi and Milios, 2013, Milios, 2013, Organic resources and biological treatment, 2010).

For the substitution of plastic materials analyzed in this study, the four more demanded plastics in Europe are selected: low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP) and poly-vinyl chloride (PVC). These polymers represent almost 60% of the European plastics net consumption (Plastics Europe, 2015). LDPE is used in food packaging and in light and flexible products like plastics bags; HDPE is used in the production of more resistant materials like house pipes or plastic toys; PP is present in durable products like car bumpers or office folders; finally, PVC can be found in window frames or wellington boots (Plastics Europe, 2015).

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 total products, see Equation 1. E_{BAU} is calculated from emissions associated with landfilling and incineration (waste management) and the impact of the background processes (emission factors, see Table 3) for transportation fossil fuels (EF_{fuel}) and plastics ($EF_{PE/PP}$ and EF_{PVC}). $E_{landfill}$ represents the GHG emissions from the landfill, which depend on the behavior of landfilled materials. Although IPCC and EPA establish default values, some parameters are country-specific and even vary between regions (see the Supplementary Material

(SM)) (EPA, 2014, IPCC 2006). $E_{incineration}$ represents the GHG emissions from MSW refuse incineration with energy recovery (see the SM). The current EF value for fossil transportation fuels in Europe is used even though it is expected to increase in the short term (COM/2016/0767, Directive 2015/1513).

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

3.3. Modeling of the BIO system

For the production of renewable-derived drop-in plastics, the MSW refuse has to be pretreated and converted into refuse derived fuel (RDF) which is then further processed in the thermochemical biorefinery (Aracil et al., 2017). This pretreatment can vary depending on the characteristics of the MSW refuse. In spite of this, RDF production increases the lower heating value (LHV) of MSW refuse up to around 16 MJ kg^{-1} (Arena et al., 2015, Onel et al., 2014, Patel et al., 2012, Yassin et al., 2009). Like for other assessments of biomass residues, there are no upstream emissions (e.g. associated with cultivation, harvesting and direct and indirect land-use change) to be allocated to the MSW refuse (Directive 2015/1513).

The production of drop-in chemicals from RDF is modeled according to a previous work of the authors where a thermochemical biorefinery producing dimethyl ether (DME) and methyl acetate (MA) from lignocellulosic biomass was technically, economically and environmentally assessed (Haro et al., 2015a, Haro et al., 2013b). In this study (Figure 3), DME is used as a substitute for fossil diesel and as a drop-in chemical for plastics production (PP and PE). It is assumed for a base case scenario that half of the DME produced in the biorefinery will be used as a fuel and the other half as a drop-in chemical. MA is used as a drop-in chemical for the production of PVC. The conversion efficiency from chemical to plastic (mass basis) and the energy and material balance of the biorefinery, which also produces biofuels and heat and/or electricity, are shown in Table 2. The share of LDPE and HDPE is assumed to be equal to the demand of LDPE and HDPE in Europe (see Table S5 in the SM).

For the incorporation of Bio-CCS, the capture efficiency is assumed to be 25% corresponding to the amount of pure CO_2 that needs to be captured in the biorefinery (pre-combustion CO_2 capture) to guarantee proper functioning of the downstream catalytic synthesis (Haro et al., 2013b). The capture efficiency is defined as the carbon captured in

Bio-CCS relative to the total carbon emitted by the biorefinery. However, larger fractions of carbon capture are possible if the flue gases are also treated (post-combustion CO_2 capture).

Table 2. Energy and material balance of the thermochemical biorefinery producing chemicals from MSW refuse and conversion efficiencies (LHV basis) from drop-in chemicals to renewable-derived plastics (adapted from Haro et al., 2013a, Haro et al., 2013c).

Input (Biorefinery)		**Biorefinery**		**Output (biorefinery)**		**Conversion efficiency from drop-in chemical (p_i) (LHV basis)**	
RDF (MW_{th})	100	**Net efficiency (LHV basis)**	41%	**DME (Mt/yr)**	24.1	p_{PE}	39%
RDF (t/h)	22.5	**Electricity consumption in RDF production (GWh/yr)**	8.2	**Methyl acetate (Mt/yr)**	14.9	p_{PP}	34%
LHV (MJ/kg)	16	**Bio-CCS (t of carbon/h)**	1.5	**Net electricity (GWh/yr)**	57.1	p_{PVC}	67%
Total (TJ/yr)	2,880	**Capture efficiency in Bio-CCS (pre-combustion)**	25%	**Total (TJ/yr)**	1,173		

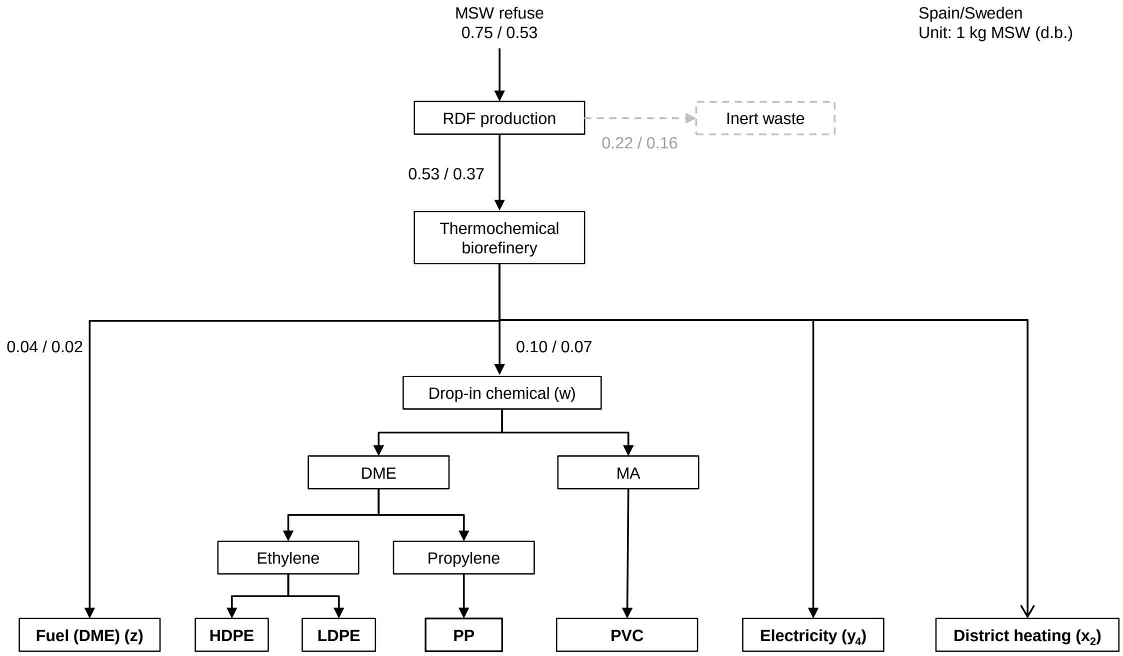

Figure 3. Description of the thermochemical biorefinery as a part of the BIO system. Final products are shown in bold. Values are in dry mass basis considering 1 kg of MSW as input for Spain and Sweden. The conversion efficiencies for renewable-derived plastics are in Table 2 and the shares of renewable-derived plastics, fuel, electricity and heat (district heating) in Table 4.

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

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

The GHG balance of the biorefinery (E_{TB}) measures the average cradle-to-grave GHG emissions, in g of CO_2 eq. MJ^{-1} of total products, associated with the life cycle of the products from the thermochemical biorefinery. A methodology for the GHG balance in the production of biofuels was previously published based on an attributional LCA, and using the standard LCA characterization method, GWP(100), and characterization factors defined by IPCC AR5 (Aracil et al., 2017, Haro et al., 2015a). In this study, the production of plastics from renewable-derived drop-in chemicals entails some changes in the cradle-to-grave carbon flows (e) compared to the previous work:

- In the GHG emissions from the thermochemical biorefinery (e_p), the anthropogenic emissions in the transformation of the drop-in chemical into renewable-derived plastics are incorporated.
- The average biogenic carbon storage in renewable-derived plastic materials ($\bar{e}_{pool}$) is introduced in the equation as a negative term since it represents a GHG removal from the atmosphere (see Appendix A for the modeling of average biogenic carbon storage in renewable-derived plastics).

Figure 4 shows the carbon flows and pools considered in the calculation of E_{TB}. The carbon flows are emissions to the atmosphere and the carbon pools are the two alternatives of carbon storage considered in this study (renewable-derived plastics and Bio-CCS). The biogenic emissions are CO_2 emissions from the biogenic fraction of the MSW refuse (usually more than 50% (IEA, 2012)) and anthropogenic emissions are fossil emissions and non-CO_2 GHG emissions from the biogenic fraction. For more information, see the SM.

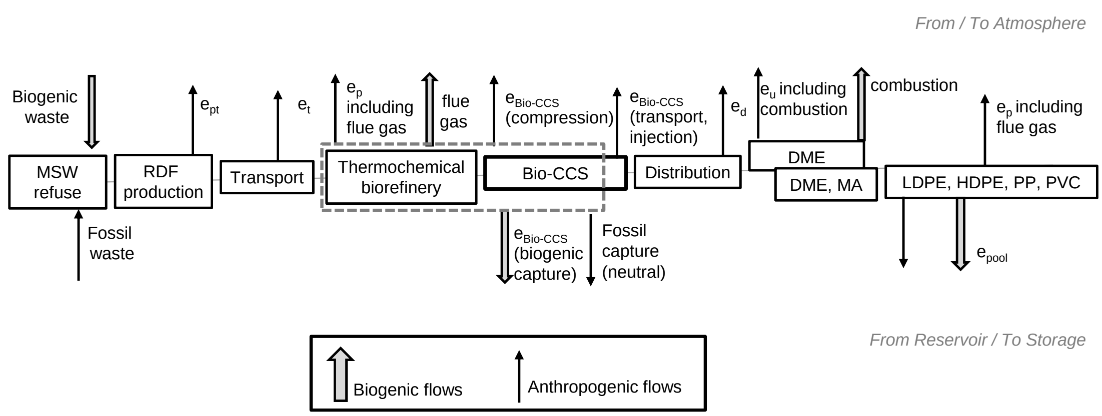

Figure 4. Carbon flows (biogenic and anthropogenic) in the production of renewable-derived plastics from MSW refuse.

Figure 5 shows the carbon balance in the production of renewable-derived plastics from MSW refuse. Around a third of the total carbon accumulated in MSW refuse is stored in the final products (fuels and renewable derived plastics) whereas the rest of the carbon is emitted in the flue gas. Considering CCS incorporation to the plant, an additional 17% of the total carbon would be captured. Therefore, just over 50% of the carbon is emitted to the atmosphere during the processing. Therefore, there is a fraction of the biogenic carbon in biofuels, in renewable derived plastics, in captured CO_2 in the CCS process and in the

CO_2 emitted to the atmosphere. The biogenic carbon stored in fuels is totally emitted to the atmosphere after it has been used (combustion) whereas there is a variable biogenic carbon stored in renewable-derived plastics. When the renewable-derived plastics become waste, they can be recycled, incinerated or landfilled. If they are recycled, the carbon remains stored for at least one more life cycle. However, after a number of recycling cycles the plastic material will be eventually incinerated or landfilled. In the case of incineration with energy recovery, the carbon stored is totally emitted to the atmosphere whereas in the case of the landfilling, the carbon is permanently stored since degradability of plastic waste is very low. Hence, there are three types of carbon storage: carbon stored through the Bio-CCS process, carbon stored in plastics still in use in the market (original and recycled plastics) and carbon stored in plastic waste landfilled. The fraction of total carbon stored after 100 years is higher in the case of Spain (8.0%), where landfilling is dominant, than in the case of Sweden (2.3%), where landfilling is negligible. The biogenic carbon stored by Bio-CCS incorporation would be about twice as the biogenic carbon stored in the renewable-derived plastics in the Spanish case whereas seven times in the Swedish one.

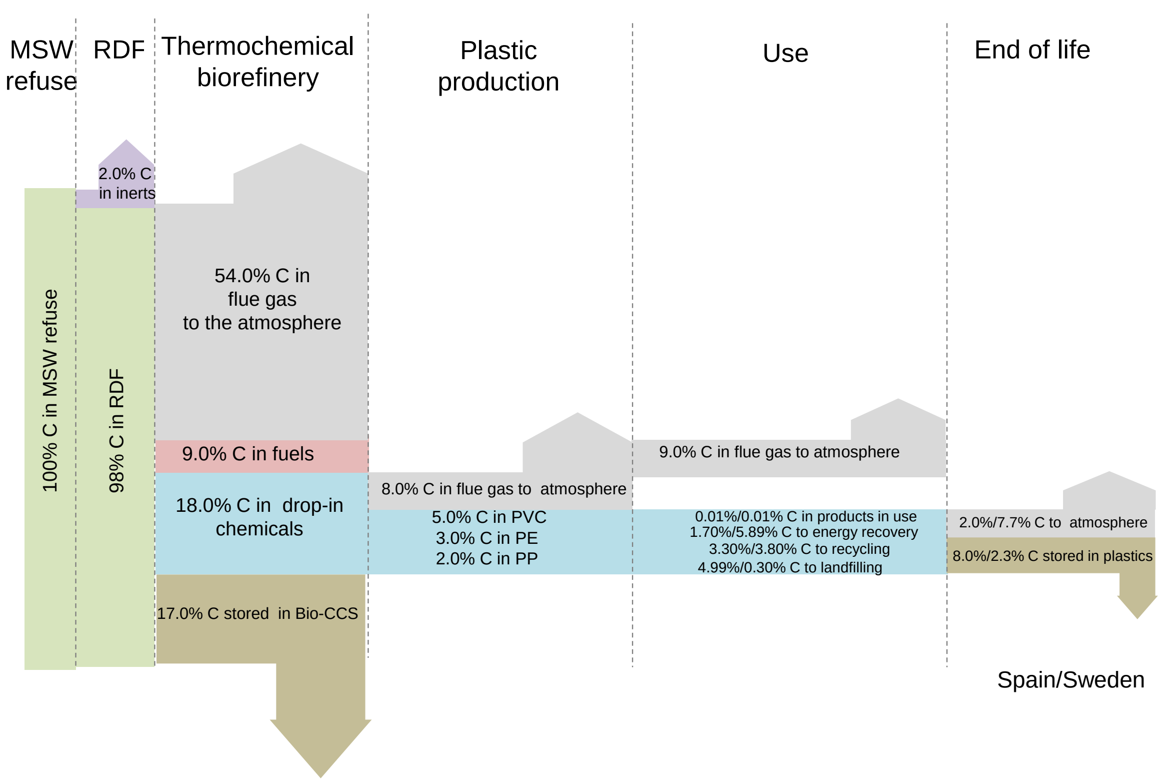

Figure 5. Carbon balance in the production of renewable-derived plastics from MSW refuse in Spain and Sweden.

For the electricity balance, the EF (Table 3) depends on the selected country and, therefore, the average values from the grid mix are used (EUR 27215 EN, 2015). For the heat balance, a conservative value 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 (Systems Perspectives on Biorefineries, 2014). There is no heat demand in the Spanish case.

3.3.1. Dynamic modeling of the biogenic carbon storage in renewable derived-plastics

The climate impact of the temporary storage of biogenic carbon in woody structures has been described in some LCA studies (EUR 24708 EN, Pawelzick et al., 2013). Considering renewable-derived plastics, the storage of biogenic carbon depends on the type of plastic material and its disposal. Whereas landfilled plastic waste will form, for all intents and purposes in this study, a permanent carbon pool, plastics waste recycled will be a temporary carbon pool whose permanence will depend on the lifetime of the plastic material and the number of recycling cycles achieved. In the case of energy recovery, there is only a carbon flow to the atmosphere. For the selected plastic materials, LDPE and HDPE are used in the production of short-time products whose lifetime is under 1 year, e.g. plastic bottles. However, PP and PVC present a longer durability and resistance and they are considered to be used mostly in the production of long-life products (over 1 year). In Europe, 60% of plastics materials are long-life plastics whereas a 40% are short-life plastics. Moreover, in 2012, a 44% of plastics became waste whereas 56% of them were still in use (Plastics Europe, 2015). Therefore, applying Eq. A1, 6.7% of long-life plastics and 100% of short-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 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 (Eurostat, Plastics Europe, 2015). Recycled plastics retain an important fraction of the carbon stored in the original plastic material. Plastic waste going to incineration releases the carbon stored as both CO_2 and non-CO_2 GHG to the atmosphere (IPCC, 2006). Plastic waste going to landfill disposal retains all the carbon (Plastics Europe, Plastics Europe, 2015). Since two alternative schemes of MSW management are considered for the biogenic carbon in MSW refuse (BIO and BAU), the differential result would cancel out the absorption of CO_2 in the original biomass (within the MSW refuse). Emissions of biogenic CO_2 are considered for all the systems but they are also largely in balance cancel out the between the BIO and BAU systems, except for the differences in the amounts of carbon stored in landfills, plastic products still in use and recycled plastics.

In order to calculate the equivalent storage of biogenic carbon in 1 MJ of renewable-derived plastic produced in one year (e_{pool}), three parameters are defined (Figure 6): 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}). In Figure 7, the evolution of each parameter from a single carbon input in the year 0 is shown (i.e., not a sustained production). 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.

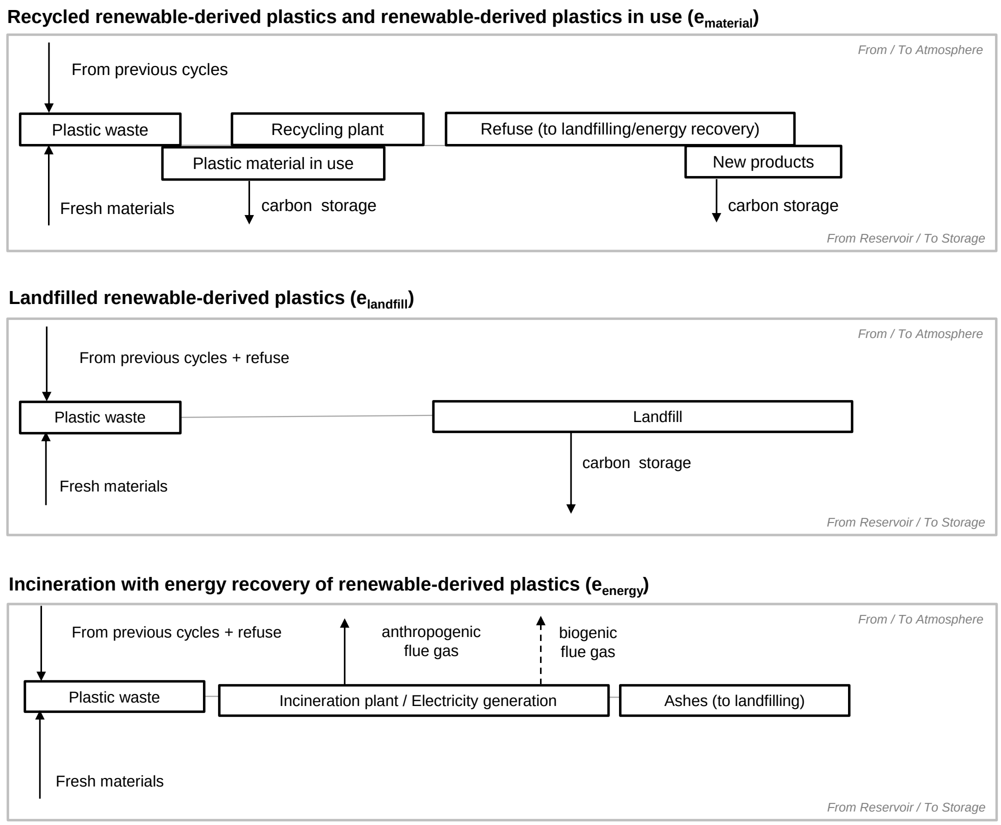

Figure 6. Representation of the three parameters defined to calculate the dynamic biogenic carbon storage in renewable-derived plastics.

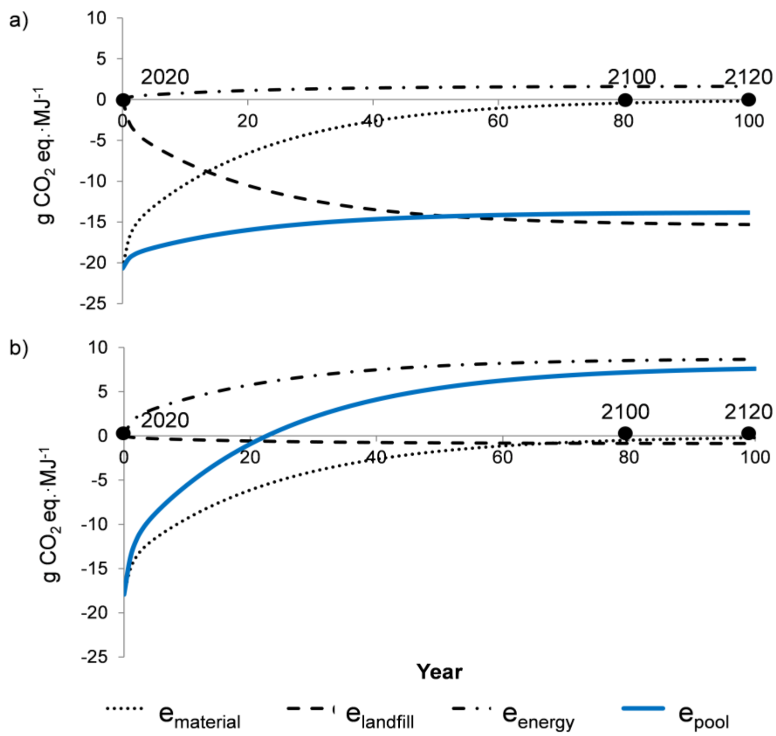

Figure 7. Evolution of the biogenic carbon pool for a single 1 MJ of renewable-derived plastic produced in year 0 (e_{pool}), as a function of the carbon flow (e_{energy}) and pools ($e_{landfill}$ and $e_{material}$) for 100 years in g of CO_2 eq.·MJ^{-1} of renewable-derived plastics. For the representation specific data for Spanish (a) and Swedish (b) waste management schemes are used.

3.4. Differential GHG impact (BIO-BAU)

The differential GHG impact in the case of using MSW refuse as feedstock (Equation 3) determines whether climate change mitigation would be higher if the fraction of the MSW went to landfilling and incineration or to the thermochemical biorefinery. The differential GHG impact is expressed in terms of g CO_2 eq.·MJ^{-1} of output for a 100 years period. For more information see the SM.

Differential GHG balance = E_{BIO} - E_{BAU} (3)

The functions represented need to be the same in all the systems considered. In order to compensate for the deficit in energy functions (i.e., heat and electricity) in the BIO system and the substitution of transportation fuels and plastics in the BAU system, we use background processes (emission factors). For the calculation of the differential GHG impact, the BAU system is considered to be unchanged for the 100 years period (static model). Table 3 and Table 4 present the emission factors and the shares of renewable-derived plastics, drop-in chemicals, fuels, heat and electricity in the BAU and BIO systems, respectively.

Table 3. Emission factors for transportation fuel and electricity production (biogenic-CO_2 emissions are excluded).

Emission factors (g CO_2 eq.·MJ^{-1})	**Spain**	**Sweden**
BAU system		
EF_{fuel} (Directive 2015/1513)	90.3	
$EF_{PE/PP}$ (Plastics Europe, 2014)	38	
EF_{PVC} (ECVM and Plastics Europe, 2005)	94	
BIO system		
$EF_{electricity}$ (EUR 27215 EN, 2015)	110.7	17.5
EF_{heat} (Systems Perpectives on Biorefineries, 2014)	-	-68

Table 4. Shares of renewable-derived plastics, drop-in chemicals, fuels, heat and electricity in the BAU and BIO systems (LHV basis), as defined in Figure 2 and 4.

Share (LHV basis)	**Spain**	**Sweden**
w	24%	13%
x_1	0%	54%
x_2	0%	10%
x_3	0%	44%
y_1	14%	0%
y_2	39%	21%
y_3	40%	13%
y_4	13%	8%
z	23%	12%

3.5. Time-dependent assessment of GHG emissions

Unlike in the differential 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 (COM (2014) 397 final Directive 1999/31/EC, Williams, 2016). The analyzed evolution assumes a progressive banning of landfilling in Europe, a decarbonization of the electricity sector and an increase in the emissions from fossil transportation fuels. 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 (State Framework Plan for waste management 2015-2020). From the year 2025 to 2120, the management is considered to be split into: 1% of landfilling, 65% of recycling and 34% of energy recovery. As Sweden is closer to the European targets than Spain, an objective 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 takes into account the carbon stored in plastic materials (see Figure S1 and S2 in the SM). 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 reach 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. Absolute Global Warming Potential (AGWP) is a cumulative formulation keeping the GHG emission memory from early years, whereas Absolute Global surface Temperature change Potential (AGTP) provides an indication of the actual surface temperature change at a given time after a GHG emission. AGTP is considered to be more transparent than AGWP (Guest et al., 2013b, Guest and Strømman, 2014). Therefore, in this study the AGTP parameter is used. To compare different regions where the feedstock composition differs, the biogenic emissions cannot be modeled. 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 ($e_{Bio\text{-}CCS}$) are modeled as negative contributions. Only Well Mixed GHG (WMGHG) including CO_2, CH_4 and N_2O are considered. The values of AGTP for BIO and BAU systems are calculated from the annual emissions of each WMGHG (see the SM). The Differential Climate Impact (DCI) is based on AGTP values (Equation 4) giving a direct comparison of the climate benefit in the considered region at a specific time.

$$\text{Differential climate impact} = AGTP_{BIO} - AGTP_{BAU} \quad (4)$$

4. Results and discussion

4.1. Differential GHG impact (BIO-BAU)

The differential GHG impact does not consider the evolution of MSW management in Europe nor the dynamic storage of biogenic carbon in renewable-derived plastics (i.e., only the average biogenic storage is considered assuming current waste management, $\bar{e}_{pool}$). Thus, it cannot give an accurate comparison of the two GGR technologies (renewable-derived plastics and Bio-CCS). However, it serves to understand the behavior of the BAU and BIO system and the relative impact of the main parameters on the study. Prior to directly compare the production of renewable-derived plastics and the Bio-CCS incorporation, the emissions in the BAU and BIO system are presented (Figure 8). The emissions associated with the BIO system (E_{BIO}) are higher in Spain than in Sweden (68 and 15 g CO_2 eq.·MJ^{-1}, respectively). Therefore, the balance of electricity and heat has a larger impact than the higher biogenic fraction of MSW refuse in Spain. Sweden has a very low-carbon electricity grid and heat mix. Conversely, the comparison of the emission associated with the BAU system (E_{BAU}) (134 and 183 g CO_2 eq.·MJ^{-1} for Spain and Sweden, respectively) reveals the impact of the larger biogenic fraction in Spain and high landfilling rate. It is clear that there is little impact of renewable-derived plastics (-7 and +1 g CO_2 eq.·MJ^{-1} for Spain and Sweden, respectively –orange dotted area in Figure 8–). The $\bar{e}_{pool}$ is positive for Sweden due to the low biogenic fraction in MSW refuse and the high incineration rate. The impact of renewable-derived plastics production is less significant than for Bio-CCS (-27 and -28 g CO_2 eq.·MJ^{-1} for Spain and Sweden, respectively –blue dotted area in Figure 8–). Therefore, Bio-CCS would seem to be a more appropriate GGR technology using a static assessment. Anyway, the differential GHG impact is negative in all cases, i.e., there is a climate benefit considering a static assessment.

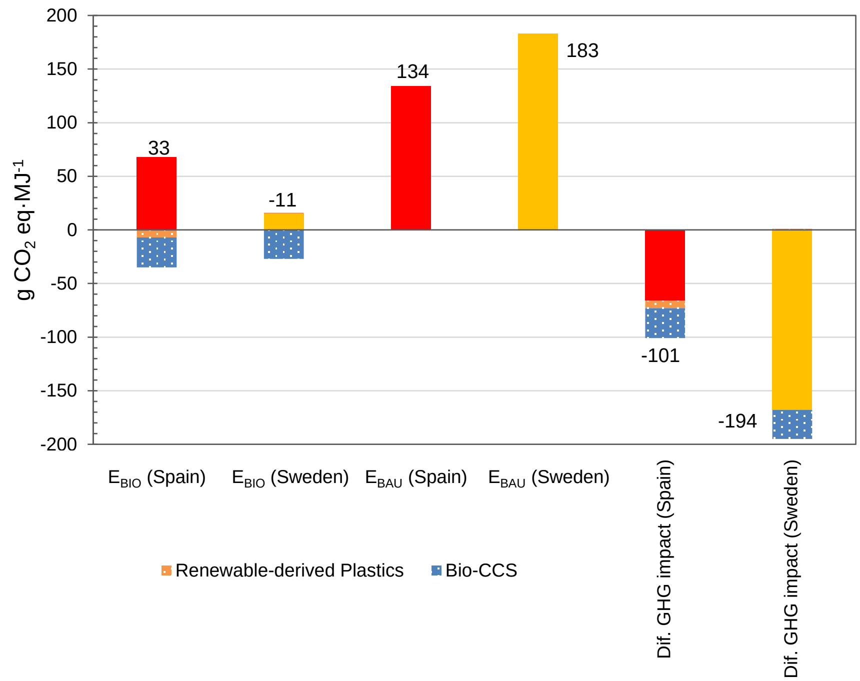

Figure 8. Differential GHG impact based on the emissions from the BIO and BAU systems. The impact of renewable-derived plastics and Bio-CCS is disaggregated. The numbers refer to net values including both renewable-derived plastics and Bio-CCS.

4.2. Differential climate impact (DCI)

For a comprehensive analysis of the production of renewable-derived plastics as a GGR technology, the DCI is an appropriate indicator since it includes the dynamic biogenic carbon storage alongside with the evolution in the BAU system. For a first comparison of the production of renewable-derived plastics and Bio-CCS incorporation, it is convenient not to combine both GGR technologies into the same system. Therefore, Figure 9 shows the DCI of the production of renewable-derived plastics (solid orange line) as modeled in this study along with the DCI of Bio-CCS incorporation into a BIO system producing transportation fuels and electricity from MSW refuse (solid blue line) and the DCI of the same system without any GGR technology (i.e., producing transportation fuels and electricity from MSW refuse, dashed grey line). The last two DCI were calculated in a previous study (Aracil et al., 2017). In Figure 9, the x-axis corresponds to the GHG emissions of the BAU system. Therefore, if the lines are over the x-axis, there is a climate worsening with respect to BAU system, but if they are below the x-axis, a climate benefit is achieved. Moreover, if the lines are within the shaped green area, it means that the climate benefit involves a lower warming than for the BAU system. Below the shaped green area, a net cooling of the atmosphere is achieved. In both countries, there is a climate benefit after the 100 years period, i.e., there is a negative DCI, considering either renewable-derived plastic production or Bio-CCS incorporation. The trend of the DCI curves is similar for all cases, which reflects the large impact of the 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, which is also lower than for the BIO system without any GGR technology. Therefore, both GGR technologies are comparable in the medium term. In Spain, there is a minimum for the DCI in year 17 because of the impact of avoided emissions (especially methane) from the landfill (Cherubini et al., 2008, Clausen and Pretz, 2016, Fiorentino et al., 2015). However, since methane emissions have a short term impact on climate change, the DCI of plastic production is reduced after the first 30 years. This increase is later balanced since the landfill share is drastically reduced over time, similarly as previously indicated in biofuel production (Aracil et al., 2017). The increase of the incineration share has a negative impact on Spanish results, making 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. In Sweden, since the incineration share decreases over time, there is a larger storage of biogenic carbon in plastic materials. Hence, the production of renewable-derived plastics compares favorably with Bio-CCS incorporation in the long term (year 2120), which could not have been anticipated by the differential GHG impact.

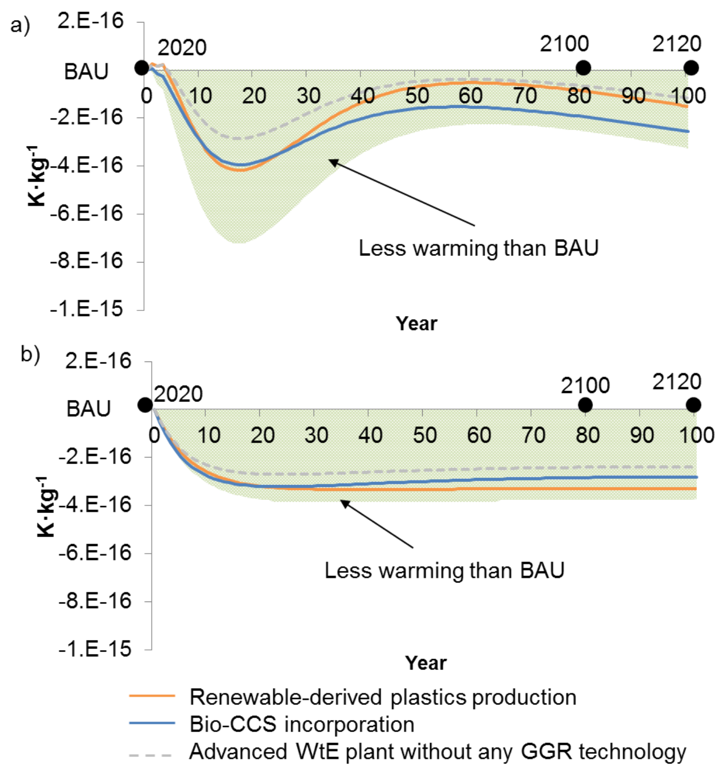

Figure 9. Differential climate impact for the sustained production of renewable-derived plastics from MSW refuse, Bio-CCS incorporation to an advanced WtE plant producing transportation fuels and electricity and an advanced WtE plant producing transportation fuels and electricity without any GGR technology in Spain (a) and Sweden (b).

To understand the potential of renewable-derived plastic production compared with Bio-CCS incorporation in the same advanced WtE plant, a sensitivity analysis is performed based on the capture efficiency (Bio-CCS) and the ratio of DME (drop-in chemical) used in the production of plastics in the biorefinery. Figure 10 presents the results, where the base case assumes an advanced WtE plant incorporating both renewable-derived plastics production and Bio-CCS. For the base case, the fraction of DME used for the production of plastics is assumed to be 50% and the fraction of carbon captured by the Bio-CCS is 25% (reflecting actual requirements for syngas conditioning for drop-in chemical synthesis (Haro et al., 2013b)). As commented before, the trend of the curves is similar for both GGR technologies, although now the sensitivity of the DCI for the renewable-derived plastics is reduced compared with the Bio-CCS. Only for the first 30 years in Spain, there is a comparable impact of the share of plastic production and capture efficiency. A plastic share of 100% would be equivalent to increasing the capture efficiency 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 fraction of captured carbon 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 use of more or less DME in the production of plastics (instead of being used as a transportation fuel) depends on the market and does not involve any technical constraint. However, a higher capture efficiency (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) (Chen et al., 2006) and would increase the process complexity (Aracil et al., 2017, Oreggioni et al., 2017). Therefore, it is not likely to have larger capture efficiency for Spain.

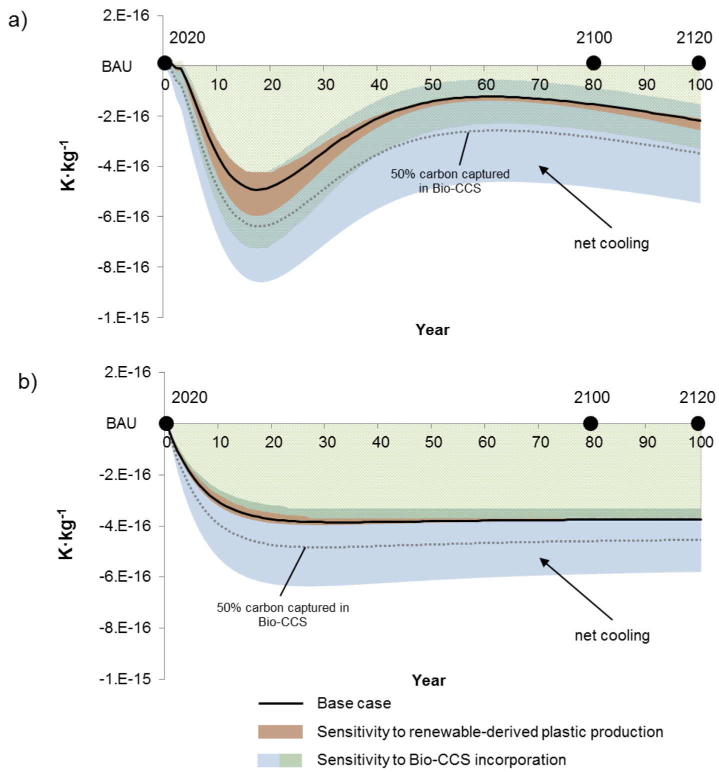

Figure 10. Sensitivity analysis of the DCI to the share of drop-in chemical used for renewable-derived plastic production and the capture efficiency (Bio-CCS) in the advanced WtE plant for Spain and Sweden.

4.3. Potential of renewable-derived plastics versus Bio-CCS

As shown above, the potential of carbon capture using renewable-derived plastics is similar to the one of Bio-CCS incorporation. Unlike other GGR technologies, the specific climate benefit of renewable-derived plastics and Bio-CCS does not involve a response of the earth system to their implementation (Smith et al., 2016). 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 but expensive, albeit CO_2 storage still needs further experience at scale: assessing, conditioning and controlling geological reservoirs for the long-term storage and the risk of uncontrolled leakages). Besides, the lack of understanding and acceptance of the technology by the public and some stakeholders also contributes to delays in CCS deployment (IEA, 2013).

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 plastics types and possible applications so that they become functionally equivalent to fossil-derived plastics (Plastic waste in the environment, 2011). 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.

Recently, Fridahl 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 (Fridahl, 2017). 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 the IAMs, should be assessed in attempts to meet the 2 ºC goal. The results shown in this paper concerning the climate benefit of renewable-derived plastics as an alternative GGR technology are promising. 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 paper need to be complemented with its 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.

5. Conclusions

The production of renewable-derived plastics proves to be an effective technology for greenhouse gas removal similar to bioenergy with carbon capture and storage. 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 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 2 ºC target.

Acknowledgements

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Appendix A. Modeling of biogenic carbon in renewable-derived plastics

For the modeling of biogenic carbon in renewable-derived plastics, products still in use in the market (P) and plastics becoming waste (W) are distinguished. Short-life plastic products ($P_{short\text{-}life\ plastics}$) are considered to become waste in less than a year. However, most of long-life plastics usually remain in the market for more than one year. Therefore, Equation A1 gives the fraction of the long-life plastic products becoming waste ($W_{long\text{-}life\ plastics}$) in one year. Long-life plastic products still in use ($P_{long\text{-}life\ plastics}$) are 1-$W_{long\text{-}life\ plastics}$.

$$W_{long-life\ plastics} = \frac{W - P_{short-life\ plastics}}{P_{long-life\ plastics}} \qquad (A1)$$

The horizon for this study is 100 years as recommended by the Intergovernmental Panel on Climate Change (IPCC) (IPCC, 2014). In the year 0, $e_{material}$ represents the input of new plastic products in the market in terms of g CO_2 eq.·MJ^{-1} of plastics (C input) whereas $e_{landfill}$ and e_{energy} have a zero value. From year 1 to 100, $e_{landfill}$ and e_{energy} increase since more and more plastic waste is landfilled or energy recovered and, therefore, $e_{material}$ decreases. Equations A2-A5 show the calculations:

- Year 0:

$$e_{material} = C\ input \cdot \alpha \qquad (A2)$$

where α is the biogenic fraction of the MSW refuse (%)

- Successive years for short-life plastic materials:

$$e_{material\ (year\ i)} = e_{material\ (year\ i-1)} \cdot W_{short-life\ plastics} \cdot r_{recycling} \qquad (A3)$$

The parameter $e_{landfill\ (year\ i)}$ is calculated as $e_{material\ (year\ i)}$ replacing $r_{recycling}$ by $r_{landfilling}$.

$$e_{energy\ (year\ i)} = e_{material\ (year\ i-1)} \cdot \frac{\beta}{\alpha} \cdot W_{short-life\ plastics} \cdot r_{energy\ recovery} \qquad (A4)$$

- Successive years for long-life plastic materials:

$$e_{material\ (year\ i)} = e_{material\ (year\ \ i-1)} \cdot (1 + W_{long-life\ plastics} \cdot (r_{recycling} - 1)) \quad \text{(A5)}$$

The other two equations ($e_{landfill}$ and e_{energy}) differ from those of short-life plastic materials changing $W_{short\text{-}life\ plastics}$ by $W_{long\text{-}life\ plastics}$. Finally, the parameter e_{pool} is calculated (Equation A6).

$$e_{pool\ (year\ i)} = (e_{material} + e_{landfill} + e_{energy})_{year\ i} \quad \text{(A6)}$$

Equation A7 is used to calculate the average biogenic carbon storage in renewable-derived plastics for the differential GHG impact.

$$\overline{e_{pool}} = \frac{\sum e_{pool\ (from\ year\ 0\ to\ 100)}}{100} \quad \text{(A7)}$$

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_{mix}	emission factor from electricity grid mix (g CO_2 eq.·MJ^{-1})
$e_{landfill}$	biogenic carbon storage in landfilled plastics (g CO_2 eq.·MJ^{-1})
e_{pool}	biogenic carbon storage in plastics (g CO_2 eq.·MJ^{-1})
e_p	emissions from processing (g CO_2 eq.·MJ^{-1})
e_{pt}	emissions from pretreatment (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_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	ratio of chemical production (1 BAU system, 2 BIO system) (%)

x_i fraction of heat production (1 BAU system; 2 biorefinery, 3 heat mix in BIO system) (%)

y_i ratio of electricity production (1 landfill, 2 incineration in BAU system; 3 electricity grid, 4 biorefinery In BIO system) (%)

z_i ratio of biofuel production (1 fossil fuels in BAU system; 2 biorefinery in BIO system) (%)

Greek letters

α Biogenic fraction of MSW refuse (%)

β Anthropogenic emission factor from MSW refuse incineration (%)

$\eta_{conversion}$ Conversion efficiency from chemical to plastic (%)

Abbreviations

AGTP Absolute Global surface Temperature change Potential

AGWP Absolute Global Warming Potential

e Carbon flows

BAU Business as usual

BIO Bioenergy

BECCS/Bio-CCS Carbon capture and storage in bioenergy

DCI Differential Climate Impact

DME Dimethyl ether

$E_{BIO/BAU}$ GHG impact of the BIO or BAU system

E_{TB} Global GHG balance of the thermochemical biorefinery

EF Emission factor (fossil comparator)

EPA U.S. Environmental Protection Agency

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

P	Products still in use
PP	Polypropylene
PVC	Poly vinyl chloride
RDF	Refuse derived fuel
SM	Supplementary Material
W	Waste
WMGHG	Well Mixed GHG

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Paper V

Implementation of waste-to-energy options in landfill-dominated countries: Economic evaluation and GHG impact

In this paper, the economic and environmental impact of various Waste-to-Energy (WtE) schemes to produce electricity from MSW refuse is evaluated and compared with landfill disposal. Both incineration and gasification alternatives are considered. Gasification option includes three different configurations: 1) FBG with internal combustion engine (ICE) 2) fluidized bed gasifier (FBG) with Organic Rankine Cycle (ORC) and 3) grate gasifier with steam Rankine cycle (SRC). It is a manuscript. A previous work inspiring this paper has been published as conference proceedings in 2016[16].

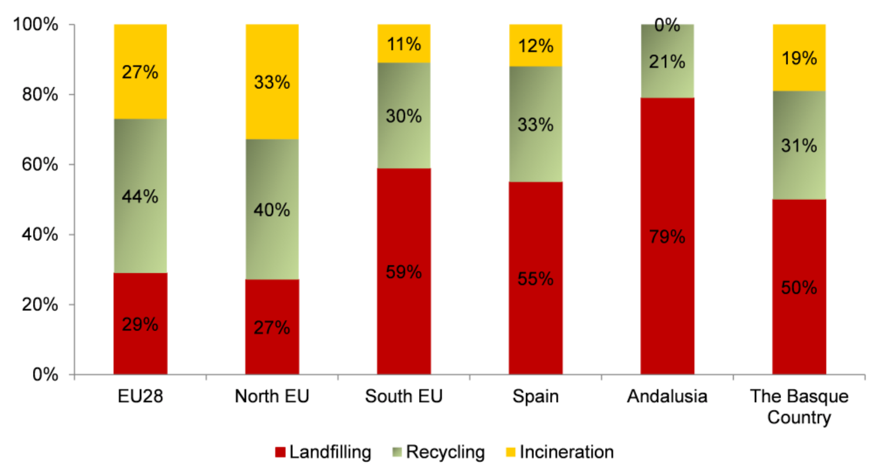
1. Introduction

Waste management remains nowadays as an environmental, technical and economic challenge. The development of integral worldwide solutions is deterred by the constraint of national, regional and local regulation. Large differences in waste management schemes are found around the world according to their level of economic development, climate conditions and historical regulation. Figure 1 (a) shows the share of recycling, incineration and landfilling for the average EU countries, and for Northern and Southern European countries (Eurostat, 2014). Landfilling requires the use of extensive land and can lead to several environmental impacts over land, atmosphere, hydrosphere and biosphere. The European Union is encouraging landfill-dominant countries to find alternative waste managing strategies, being incineration with energy recovery the most common (Eurostat, 2014). Among the management options in Europe (Figure 1 (a)), landfilling is the most popular in Southern Europe countries, since it has a lower cost compared to incineration plants. In Northern Europe, the landfilling ratio is below incineration and recycling ratios, being even negligible in some countries like Germany. The differences between regions of

[16] Aracil C, Haro P, Fuentes-Cano D and Gómez-Barea A. Implementation of waste-to-energy options in landfill-dominated countries: Economic evaluation and GHG impact.
Proceedings in http://ftp.servcbo.com/1605WASTEENG/Abstracts/Topic3B/054-spc0054.pdf

a country can be as significant as those between different countries, as it comes from the differences between The Basque Country (North Spain) and Andalusia (South Spain) shown in Figure1 (a). Although 55% of the generated MSW is landfilled in Spain every year, landfill sites are more concentrated in the South, whereas incineration plants are mostly located in North. Figure 1 (b) shows the ratios of recycling, incineration and landfilling for the Southern European countries other than Spain, which will be compared with Spain throughout this study (Italy, Portugal, Greece, Cyprus, Croatia, Malta and Romania).

Regarding the situation of incineration plants in Europe, Sweden, Denmark, Netherlands, Luxembourg and Finland achieve the highest waste incineration ratios with values ranging from 217 to 412 kg of MSW per capita (Eurostat, 2014). These countries are among the twelve countries with the highest Gross domestic product (GDP) per capita, that is, among the ten most developed countries in Europe and they are located in Northern.



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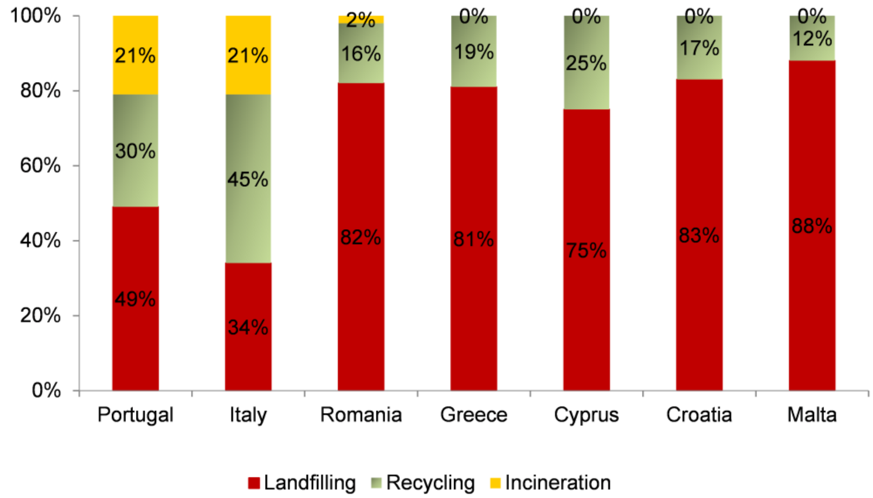


(b)

Figure 1. Current situation of MSW management in Europe and selected Spanish regions. The percentages represent the fraction of MSW incinerated, recycled or landfilled. Adapted from Eurostat (2014).

The European waste hierarchy establishes that only MSW refuse can be used as feedstock in WtE plants, and that the use of any other fraction must be justified from the life cycle thinking (Directive 98/2008). MSW refuse is the unsorted fraction of MSW that cannot be further processed (for recycling) and therefore goes into incineration or landfilling. This MSW refuse comes either from separated waste collection (as the non-recyclable fraction) or from mechanical and biological treatment (MBT) plants (a fraction separated from the compostable fraction). The MSW refuse from a MBT plant is a heterogeneous and site-specific stream although analysis of various refuse fractions from different configurations MBT plants in Spain showed that all of them fulfilled requirements for standardized solid recovered fuel (SRF) (Edo-Alcón et al., 2016, EN 15357:2012). In Europe, the SRF/Refuse Derived Fuel (RDF) production and consumption have been increasing over the last years, reaching the highest values in Northern Europe. There is also a great potential to produce RDF in Southern Europe such as in Spain, where only 1 Mt of SRF/RDF (but the production potential estimated is higher than 6 Mt per year) is produced and consumed in the cement industry and incinerators (mainly fluidized bed reactors) (ISR, 2006, AEVERSU, 2016).

A rational transformation of the waste management scheme in Southern Europe requires understanding the WtE schemes and the inherent differences between the North and the South. In Central and Northern Europe, medium or lower scale CHP plants (up to 50 kt RDF/y) can be efficiently implemented provided the heat demand is high enough. These CHP plants are mainly optimized to satisfy the heat demand with minimum costs, being electricity share generally low, typically below 10% (EUR 26729 EN). However, in Southern Europe, electricity is the desired product in CHP plants, since heat demand is generally low, so high electrical efficiency is required. To produce electricity with a reasonable efficiency (>20%) large scale is necessary (>0.5 Gt RDF/y) (Consonni and Viganò, 2012). Therefore, large scale WtE plants are required in Southern Europe, meaning that current MTB plants generating limited amount of MSW refuse (typically less than 300 kt/y (Arena et al., 2015) will need to produce and send the RDF to a centralized plant. To minimize transport costs, advanced WtE plants using gasification can be integrated with existing MBT plant. Gasification technologies can be adapted to the volume of waste available in most of existing MBT plants (medium scale, i.e. lower than 100 kt/y) to produce mainly electricity as required in Southern Europe. There is enough industrial experience in gasification plants to consider its penetration in the short and medium term. For instance, the company Energos has built eight gasification plants in Europe settled in Norway, Germany and the UK, seven of which are operational (Energos, 2017). The plants process between 30 and 78 kt RDF/year, only one producing electricity (through a Steam Rankine Cycle, SRC) and heat (CHP) and the rest producing steam for heat applications (low or medium-pressure steam). A large fluidized bed gasification (FBG) plant was came in operation in 2012 by Valmet in Lahti (Finland) processing 250 kt SRF/ y (Valmet, 2013). The produced gas is burned to produce electricity through a SRC.

This study assesses the techno-economic and GHG impact of implementing incineration and gasification-based WtE technologies in Southern European countries, using as a case study the Spanish region of Andalusia.

2. Methodology

2.1. Definition of the systems

Three systems are considered; a reference system representing current MSW refuse management system in a landfill-dominant region and two bioenergy systems where MSW refuse is used as feedstock in incineration or gasification WtE plants. Figure 2 depicts the three systems considered. The functional input unit is 1 ton of MSW refuse and the product output is electricity, to focus the analysis on regions of Southern Europe where electricity is the main product because heat demand is low, whereas the outputs y_i (i=1,2,3) are the electricity production from 1 t MSW refuse (MWh_e/t MSW refuse). The assumption to consider only electricity as valuable output is consistent with the fact that WtE plants in these regions are designed and operated to reach the so-called R1 threshold, established by EUR 26720 EN (R1 formula was amended to incorporate the climate correction factor, CCF, defined in Commission Directive 2015/1127/EU). Therefore, heat is not considered as valuable outcome in this study.

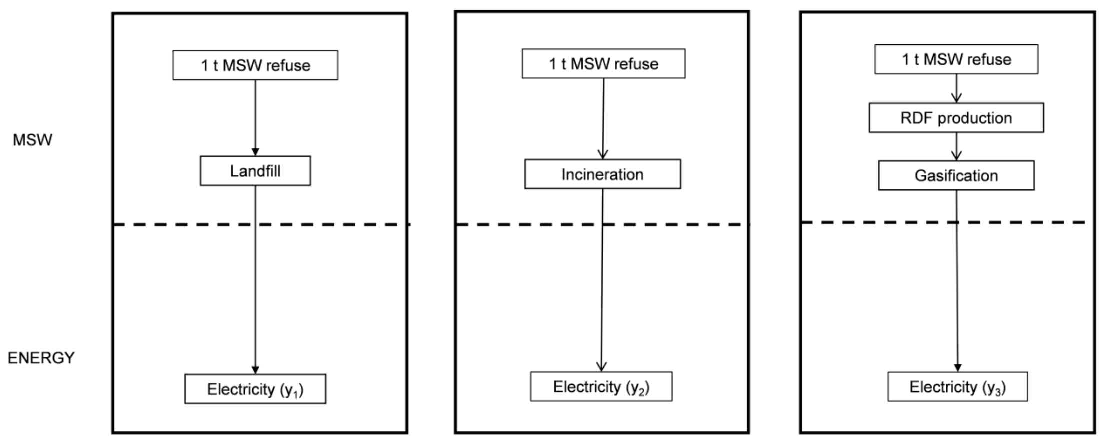


Figure 2. Definition of the reference (landfill) and bioenergy (incineration and gasification) systems in the study.

2.2. Definition of the WtE systems

Incineration is the complete combustion of the MSW refuse to produce heat that is used directly or converted into electricity in a power plant. Gasification is the partial combustion to produce a syngas, which in the context of the present study is further combusted to produce electricity. The incineration and gasification technologies considered here are:

i) Incineration system: The technology selected is a moving grate allowing the use of MSW refuse without pretreatment. This enables avoiding GHG emissions and costs associated to MSW refuse pretreatment but the heating value of the feedstock to the hearth decreases, and pollutants formation during burning increases (8-13 MJ/kg, Boesch et al., 2014; Consonni and Viganò, 2012; Yassin et al., 2009), leading to a more expensive equipment and gas treatment. A low heating value of 11 MJ/kg is considered in this study. The MSW refuse is transported from several MBT plants to a centralized large-scale incineration plant. Once defined the region under consideration, a decision of one or various incineration plants is to be made, as discussed below. Figure 3 (a) shows a simplified incineration plant diagram with a post-combustion system of syngas cleaning. The overall system efficiency is 21%, calculated assuming an overall boiler efficiency (from feedstock to steam) of 63% (Yassin et al., 2009) and a steam turbine efficiency of 33% (Consonni and Viganò, 2012). Figure 3 (b) shows the energy balance of the incineration process.

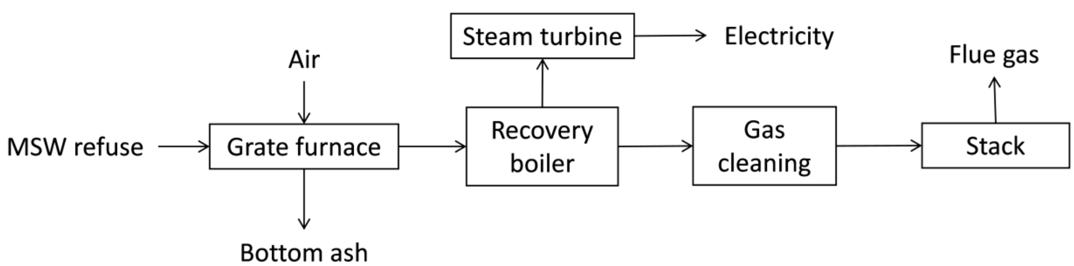
ii) Gasification system: Several technologies are considered for the production of syngas (moving grate and fluidized bed gasifiers) and to convert the syngas into electricity (internal combustion engine (ICE), organic Rankine cycle (ORC) and steam Rankine Cycle (SRC)). FB gasification technologies require a homogeneous fuel, so that the MSW refuse has to be pretreated. Moving-grate gasifiers can generally be operated with direct MSW refuse but the lower technical development of gasification compared to incineration makes to pretreat the refuse better, in order to improve the availability of the gasification process and the increase of the heating value of the feedstock to the range of 16-20 MJ/kg (Arena et al., 2015; Edo-Alcón et al., 2016; Onel et al., 2014; Yassin et al., 2009). A low heating value of 18 MJ/kg is assumed in this study. Then, in all gasification systems considered in this study the MSW refuse is converted into RDF (Figure 4 (a)). The size of the plants is adapted to the amount of MSW refuse in each MBT plant, taking advantage of the ability of gasification to produce electricity at high efficiency at small/medium-scale.

For RDF production in current MBT facilities some additional equipment is necessary. Existing MBT plants are usually equipped with various sorting for separation: manual for separate bulky items, trommel for the finest fraction, and magnetic and eddy current separators for ferrous and non-ferrous metals, respectively, as well as density separator to separate different kinds of plastics (Edo-Alcón, 2016). Additional equipment to produce high-densified RDF should incorporate shredder, dryer and pelletizer (Caputo et al., 2002a and 2002b). Heat recovery and integration is possible in schemes with RDF production by using heat from the gasifier increasing the total energy efficiency.

In the two first configurations the syngas is produced in fluidized beds. The electrical efficiency using an ICE to generate electricity is 23.8% (assuming efficiencies of 61 and 39% for gasification (cold) and engine, respectively, Di Gregorio et al., 2012). Using an

FBG in an ORC to produce electricity, the estimated electric efficiency drop to 11% (assuming a cold gas efficiency of 61% and a net electric efficiency of the ORC cycle, including the oil-gas heat exchanger, of 17.7%, Arena et al., 2015). In a GG/SRC configuration the efficiency is 13.3% (Energos, 2012) assuming a feedstock-to-steam efficiency of 44.3% and a steam turbine efficiency of 30% (Energos, 2012, Patel et al., 2012). These electrical efficiencies (from RDF to electricity) increase when RDF production is considered being the total energy efficiencies (from MSW refuse to electricity) of 31, 11 and 15% respectively (Figure 4 (b)).

Table 1 summarizes the electricity ratios (yi) resulting from the proposed schemes: incineration (y_2) and in the three gasification-based systems of Figure 2 (y_3), defined as MWh_e per t of MSW refuse. The ratio generated in the landfill (y_1) is also included and will be discussed later.



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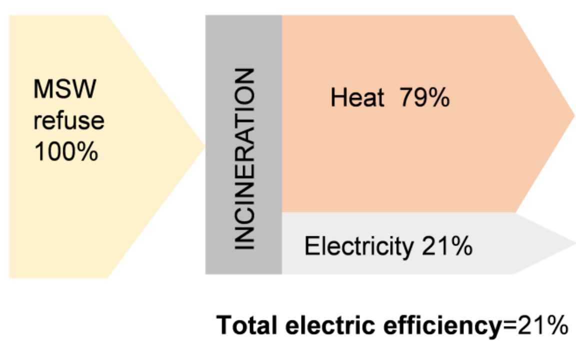
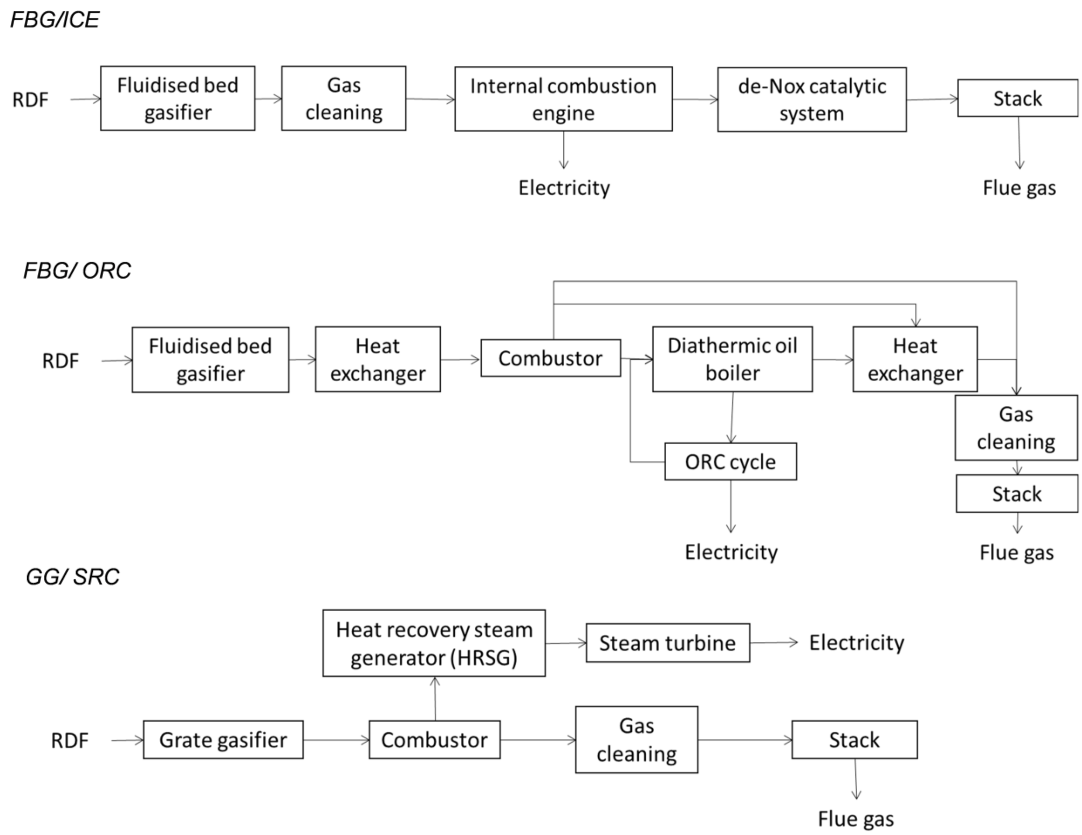


(b)

Figure 3. Simplified diagram of the incineration (a) and energy balance (based on % of energy of the input fuel) (b).


FBG/ICE
RDF
Fluidised bed gasifier
Gas cleaning
Internal combustion engine
de-Nox catalytic system
Stack
Electricity
Flue gas
FBG/ ORC
RDF
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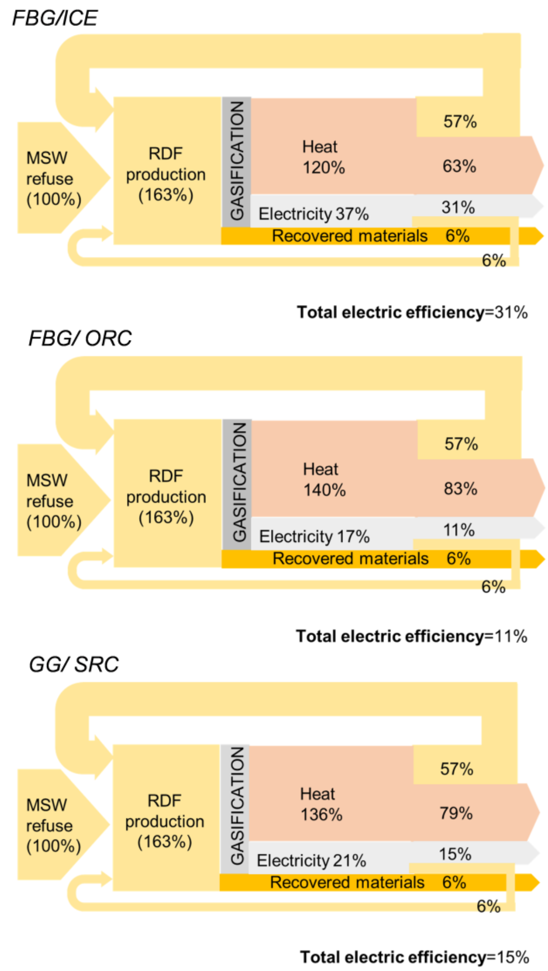
Heat exchanger
Combustor
Diathermic oil boiler
Heat exchanger
Gas cleaning
ORC cycle
Stack
Electricity
Flue gas
GG/ SRC
Heat recovery steam generator (HRSG)
Steam turbine
Electricity
RDF
Grate gasifier
Combustor
Gas cleaning
Stack
Flue gas

(a)



(b)

Figure 4. Simplified diagrams (a) and energy balances (based on % of energy of the input fuel) (b) of the WtE based on gasification plants proposed according to the gasification technology, showing the net global electric efficiency of the systems.

Table 1. Shares of electricity production using 1 ton of MSW refuse (y_i) defined as MWh_e per t of MSW refuse for the systems considered in Figure 2: landfill (1) and WtE (incineration (2) and gasification (3)) of. (Data for the estimation based on: Arena et al., 2015; Di Gregorio et al., 2012; Energos, 2012; IPCC, 2006; Patel et al., 2012; Yassin et al., 2009 as explained in the text).

	Landfill (y_1)	**Incineration (y_2)**	**Gasification (y_3)**		
			FBG/ICE	**FBG/ORC**	**GG/SRC**
MWh_e/t MSW refuse	0.18	0.64	0.9	0.34	0.46

2.3. The region of Andalusia as reference case of a dominant-landfill region

Andalusia is a Spanish region where around five millions tons of MSW are generated every year. In this region, there are no WtE plants and all the unsorted MSW is processed in MBT plants in which compostable and recyclable fractions are separated. The fraction generated at the end of the process, the so-called MSW refuse, representing about 80% of the total MSW generated per year, is landfilled, being well above the Spanish and European average. This fraction is a heterogeneous feedstock with 76% content of biomass (renewable fraction) (IEA, 2012). The Spanish Waste Framework 2016-2022 (State Framework Plan, 2016-2022) enforces: (i) the increase of energy recovery from the refuse from MBT facilities, either directly or by RDF production; (ii) the increase of the share of recovery up to 15% of MSW, either by incineration or co-incineration, and (iii) the decrease of the landfill share below 35%. It is concluded that the use of the MSW refuse as feedstock in WtE facilities is an interesting option in Spain, particularly, in regions like Andalusia. Figure 5 depicts a conceptual flowsheet with the current MSW management system in Andalusia, showing the WtE alternatives proposed in this study. This waste management system in Andalusia is similar to that of other countries in Southern Europe, like regions of Cyprus, Greece, Croatia, Malta, Romania (Figure1).

A study was carried out to estimate the production of MSW refuse of the 23 MBT plants currently running in Andalusia. The MBT plants can be considered individually in order to set up a small-scale WtE plants (gasification) o grouped to achieve higher plant energy capacities (incineration). An electricity potential of a standalone-WtE plant integrated in each MBT plant was estimated for each plant applying the efficiencies calculated for the various gasification technologies. In the case of considering the MBT plants grouped, the transport costs must be taken into account as well several criteria to find the number and size of the incineration plant for a given region. The following method is applied: (i) the incineration plants should be set up next to the MBT plants with the highest capacities; (ii) a maximum radius of 250 km is drawn around the incineration plant locations since the transport costs increase considerably beyond this distance (ISR, 2006).The MBT plants falling within the above distance feed the nearest incineration plant; (iii) a dispersion factor is used to allocate every plant according to the real distance from the incineration plant, considering 0 the location of the incineration plant and 1 the most disperse MBT plant, and assuming a linear relation with the distance for the plants between those two extremes. The size (energy potential) of the incineration plant is determined by accounting for the energy potential of the MBT plants grouped within the considered ratio; (iv) a minimum capacity of 100 MWthis considered in order to set up incineration plant with high enough electricity capacities (>20% net efficiency).

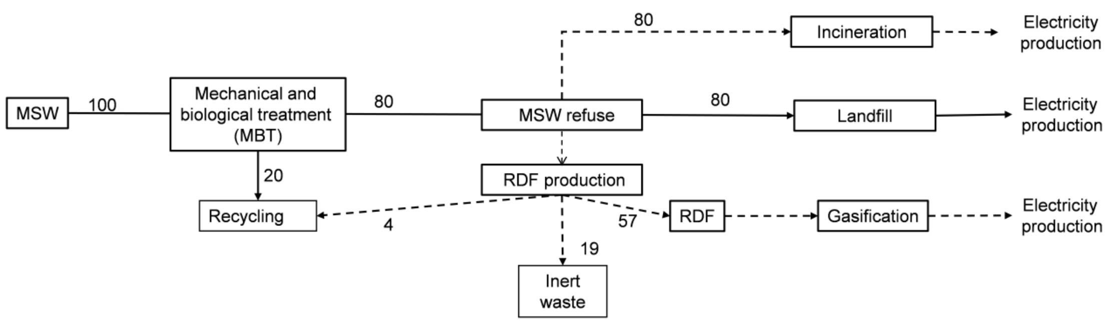


Figure 5. Current and proposed MSW management scheme in Andalusia. Dashed line shows the alternative WtE management systems proposed in this study (incineration and gasification). The values indicate the mass fraction (%) for the mass fuel input.

2.4. Techno-economic assessment

Table 2 shows the main assumptions for the techno-economic assessment. Total Investment Cost (TIC), Total Operational Cost (TOC) and Internal Rate of Return (IRR) are calculated as follow. Equipment costs are taken from literature (Arena et al., 2015; Di Gregorio and Zaccariello, 2012; Patel et al, 2012; Yassin et al., 2009) but updated to 2015 (CEPCI, 2015). The incineration plant is centralized; therefore, it has an extra cost compared with gasification plant, the MSW refuse transport from the MBT plant to the incineration plant. A cost of 20 €/t MSW refuse has been considered for a transport by truck up to 250 km of distance (ISR, 2006). In order to adapt the transport costs to the real distance between the MBT plants and the WtE plant, the dispersion factor discussed above is used. In the case of the gasification technology, TOC is calculated as the sum of fixed and variable costs. Fixed costs involve maintenance, insurance, labor and management and services. These costs are calculated as a fraction of TIC (Table 3). Variable costs entail RDF production, residues disposal and commodities consumption (Table 4). The revenue to the plant depends on the wholesale electricity tariff, the landfill gate fee, and the bottom ash and the recovered material sale (Table 5). These aspects are taken into account for the different countries and regions analyzed here. Since the landfill gate fee varies greatly from country to country (and even from region to region in the same country) the techno-economic assessment will be made by considering a range of this value and making a sensitivity analysis of the assumed values for different regions. An incineration plant has usually a lower TIC per ton of MSW refuse than gasification plant except for the case of a FBG/ORC WtE plant but TOC is usually higher in incineration (Table 6).

Table 2. Assumptions and parameters used in the techno-economic assessment.

Inflation	2%
Capital cost	5.5%
Hours/year	7500
Corporation tax	30% of taxable (incineration) 25% of taxable (gasification)
Scale factor	0.6 (Di Gregorio and Zacariello,2012)
Lang factor	3.1
Amortization years	15 years (incineration) 10 years (gasification)

As it was discussed**,** in gasification plants densified RDF is required and it is not considered in the literature used for the economic estimations made in the present study; therefore it must be added for the economic analysis of gasification plants. Table 8 gives the economic cost associated to the MBT plant modification required to produce RDF. Dryer is the most expensive equipment achieving 9.6 €/t RDF; however, the operational costs are higher in the pelletizer, almost 1 €/t RDF (Table 7) (Caputo el al., 2002; Holmberg et al., 2004).

Table 3. Fixed costs in the gasification plants (Arena et al., 2015; Haro et al., 2012).

	%TIC
Maintenance	1.5%
Insurance	0.5%
Labor	6%
Management and services	2%

Table 4. Variable costs in the gasification plants (Arena et al., 2015; Di Gregorio and Zaccariello, 2012).

	FBG/ICE GG/SRC	**FBG/ORC**	**unit**
Residues disposal	10		€/ton RDF
RDF	2		€/ton RDF
Chemicals and additives	5	8	€/ton RDF

Table 5. Data for the calculation of the revenue in incineration and gasification plants considering Andalusia.

Electricity sale (€·GJ^{-1})	**16**
Gate fee (€·t MSW^{-1})	Variable
Bottom ash sale (c€·ton bottom ash^{-1})	15 (AEVERSU, 2016)
Recovered material sale in RDF production (€·ton metals^{-1})	59 (ferrous) (ISR, 2006)
	102 (non-ferrous) (ISR, 2006)

Table 6. Calculated TIC and TOC per ton of MSW refuse in Andalusia.

	Incineration	**Gasification**		
		FBG/ICE	**FBG/ORC**	**GG/SRC**
TIC ($€_{2015}$·ton MSW refuse^{-1})	211	332	197	383
TOC (€·ton MSW refuse^{-1} ·year^{-1})	72	58	33	52

Table 7. Economic cost associated to the TMB plant modification (Caputo et al., 2002a and 2002b).

	Shredder	**Dryer**	**Pelletizer**
TIC (€/t RDF)	1.2	9.6	6.8
TOC (€/t RDF)	0.3	0.8	0.9

2.5. Method for estimation of GHG emissions

GHG balance is a stationary assessment of the cradle-to-grave GHG emissions associated to the life cycle of a process and their products (E). A methodology, initially developed by the authors to calculate the GHG balance in the production of biofuels and/or drop-in chemicals from MSW refuse (Aracil et al. 2017), has been adapted to the production of electricity in this study. The main difference for the application of GHG balance to MSW with respect to biomass is that the fossil fraction of the MSW has to be taken into account. Hence, we distinguish between biogenic and anthropogenic emissions. Biogenic emissions is the renewable fraction contained in the MSW refuse released as CO_2, having a neutral impact on climate change, whereas anthropogenic emissions is the fossil fraction in MSW refuse released as Well-Mixed GHG emissions (WMGHG). In anthropogenic emissions, biogenic non-CO_2 GHG emissions (CH_4) are also included (Aracil et al., 2017).

The Equation 1 is used to estimate the GHG emissions in the production of electricity from MSW refuse (total =processing + transport + ash treatment):

$$E_{WtE}=e_p+ e_t + e_{ash} \tag{1}$$

where

• e_p: takes into account the emissions from the WtE plant, including all process units processing MSW refuse or RDF into electricity (conversion, gas cleaning and electricity production). There are three main contributors: emissions from MSW refuse pretreatment (RDF production), the upstream emissions associated to consumables used in the process and the combustion emissions from the fossil fraction (Ecoinvent).

• e_t: takes into account the emissions from transport of the MSW refuse to the WtE plant. An emission factor of 4.1 kg CO_2/t MSW refuse was used considering a maximum distance of 250 km by truck (European Commission, 2015).

• e_{ash}: takes into account the emissions from ash treatment and disposal. It includes emissions from bottom and fly ash disposal (Boesch et al., 2014).

For the reference system (landfill) the electricity is produced from the biogas, a mixture of CH_4 and CO_2 from the degradable organic carbon (DOC) of the MSW refuse (IPCC, 2006). For GHG calculations, it has to take into account that only a fraction of the generated biogas is captured and used for electricity production or burned in flare. Therefore, GHG emissions are released from the landfill by biogas combustion (with or without energy recovery) and biogas leaking from the landfill site. The GHG balance of the reference system ($E_{landfill}$) corresponds with the climate impact of the CH_4 emissions and biogenic carbon storage. The CH_4 emissions from the landfill are calculated using the default values method from country-specific waste composition (Figure 6).

In 2006, the Intergovernmental Panel on Climate Change (IPCC) published guides for the calculation of the GHG emissions from solid waste disposal (IPCC, 2006). The U.S. Environmental Protection Agency (EPA) published in 2014 a related document (EPA, 2014), only differing in the modeling of the biogas collection efficiency (neglected in IPCC Waste Model). Whereas IPCC recommends not considering biogas collection in the landfill, the EPA recommends a range of 50% to 95% being the average 75%, i.e., 75% of the landfill gas generated is collected and routed to a control device. This parameter can vary considerably between countries. For instance, most of landfill sites in Romania or Serbia are uncontrolled landfills where biogas collection is not managed (Cailean and Teodosiu, 2016; Stanisavljevic et al., 2012). Although the EPA's values are attributable to landfills from USA (Coventry et al.,2016), some authors consider the EPA values applicable for Europe, whereas other authors consider that these values are not representative of a Southern European landfill (Cherubini et al.,2008; Zamorano et al.,2007; Eriksson et al.,2009; Aronica et al.,2009). A recent article analyzing Spanish landfills uses a biogas collection efficiency of 70% as the most favorable case (Chacartegui et al., 2015), so this conservative value is assumed as reference in the present study and its impact will be further assessed by sensitivity analysis. A correction factor should be introduced to distinguish between the biogas burned in the torch (without energy recovery) and with energy recovery. Since recommendation for this factor was not found in the literature, it is assumed here that 100% of the biogas collected is used for electricity production. Again, this conservative assumption brings total energy efficiency (LHV basis) of 7% for the landfill in the reference case. Finally, the GHG emissions associated to CH_4 were calculated using the default values from country-specific waste composition.

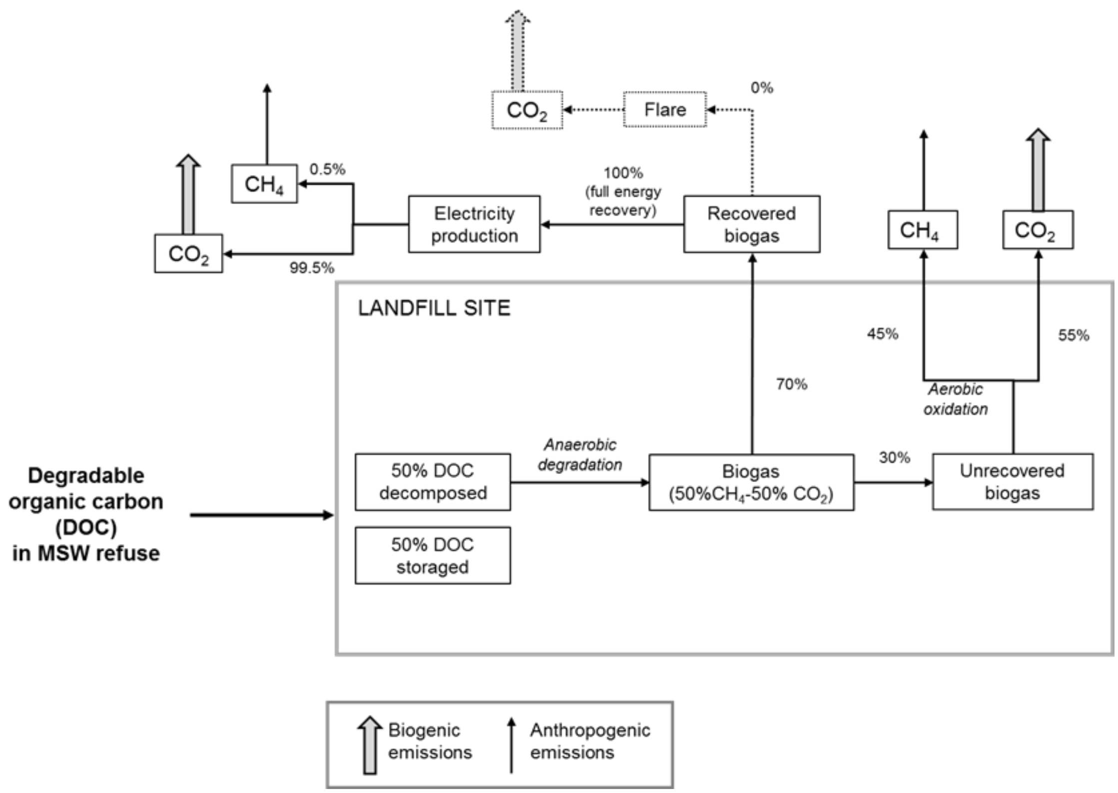


Figure 6. Emission factors considered in this study to calculate GHG emissions associated to landfilling in the reference (landfill) case (IPCC, 2016; EPA, 2014).

The differential GHG impact compares the potential reduction in GHG emissions by replacing the existing landfill scheme ($E_{landfill}$) with a WtE scheme (E_{WtE}) (Gaudreault et al.,

2015) (Equation 2), i.e., it determines how better is to process MSW refuse in WtE plants with respect to landfilling in terms of GHG emissions reduction.

$$\text{Differential GHG impact} = E_{WtE} - E_{landfill} \quad (2)$$

The GHG impact is calculated in grams of CO_2 equivalent per ton of MSW refuse. However, the landfill and WtE systems have different electricity productions per ton of MSW refuse (Table 1). Therefore, it is necessary to include in the calculation of the GHG impact the avoided fossil GHG emissions in the production of electricity. A fossil reference value for electricity production ($EF_{electricity}$) is applied, which depends on the country. Therefore, the average value from the grid mix of each country is used, which for the case of Spain is 0.398 t CO_2 eq./ MWh_e (EUR 27215 EN).

3. Results

3.1. Economic assessment

The economic comparison is shown in Figure 7, which displays the gate fee necessary to achieve 15% of internal return rate (IRR) as a function of plant fuel-input capacity (MWth), for the 4 WtE schemes considered. According to the previous discussion, incineration is considered for electricity loads higher than 100 MWth, whereas gasification schemes are displayed from small to medium capacities (5-60MWth) in order to adapt these WtE plants into existing MBT plants. As expected, the higher size plant allows lower gate fee to achieve the same economic feasibility. On the other hand, the impact of the gate fee on the feasibility is higher for gasification schemes as compared to incineration, since the reduction of the annualized cost of investment of these plants is more sensitive to the plant scale than incineration, increasing the cash flow more significantly than the wholesale electricity tariff. The FBG/ORC and FBG/IEA are seen to be better choices than GC/SRC from the economical point of view. The FBG/ORC configuration gives the lowest gate fee at a small-scale plant capacities (<35 MWth) because its lower capital and operational costs (Table 6). From 35 MWth, the FBG/ICE obtains the best results but the differences between the two technologies are very small, so it should be considered similar for the rough estimations considered in this work. However, if the gate fee was expressed as €/MWe (instead of €/t), i.e. if the fee received for 1 ton of refuse would take into account the avoided € to be paid to generate the difference of power produced by the two technologies per ton of refuse (which has to be supplied by other mean), the ICE would be much more interesting option than ORC from the economical point of view, due to the much higher electric efficiency (almost three times). However, ORC is a mature and widely spread and reliable form of energy production in CHP applications based on biomass (Turboden, 2016) with lower technical and economic requirements in gas cleaning than ICE option, so its suitability for market penetration in the short-term scenarios should be considered in terms of higher reliability.

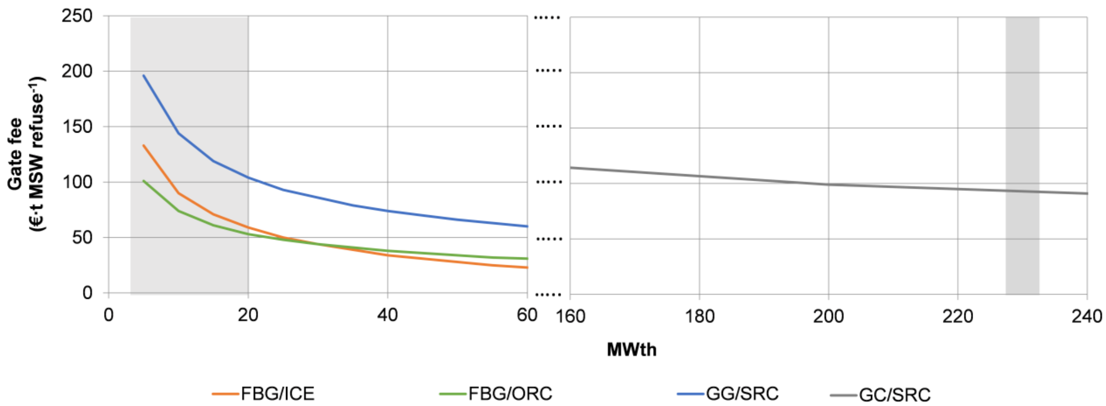


Figure 7. Economic results according to the WtE configuration (a. gasification, b. incineration) considering an IRR of 15% in Andalusia. The shaded area indicates the range of thermal energy in which the most of the MBT plants are (5-20 MW_{th}) in Andalusia (a) and the incineration plant capacities proposed in this study (b).

3.2. Environmental assessment: GHG impact

Table 9 summarizes the individual contribution of each parameter to the global GHG balance (E_{WtE}) for the WtE systems and the avoided emissions in terms of electricity production from the landfill replacement for a WtE scheme. The largest GHG emissions are associated to incineration summing up 331 kg CO_2 eq./t MSW refuse. The emissions for gasification-based schemes are considerable lower (15-25% lower than incineration): 281, 264 and 281 kg CO_2 eq./t MSW in the FBG/ICE, FBG/ORC and GG/SRC, respectively (Table 8). The main contributor to the GHG emissions is e_p, i.e. those associated to processing the MSW refuse or RDF into electricity (conversion, gas cleaning and electricity production). Considering the avoided emissions, the FBG/ICE is the best option in terms of GHG emissions per ton of MSW refuse, because of the higher efficiency compared to other options, avoiding fossil emissions. The landfilling of MSW refuse is by far the scheme with higher pollution load (454 kg CO_2 eq./t MSW refuse). Overall, the transformation of existing landfill into WtE plants leads to a net avoidance of GHG emissions from national electricity production, with the exception of the scheme based on FBG/ORC.

Table 8. Results of the GHG balance for the WtE systems and comparison with landfill in terms of avoided emissions from electricity production.

	kg CO_2 eq.·t^{-1} MSW refuse				
	GC/SRC	Gasification			Landfilling
		FBG/ICE	FBG/ORC	GG/SRC	
e_{pt}	-	(4.9)			-
e_p	324	280	270	280	
e_t	4.1	-	-	-	
e_{ash}	3.2	5.0			
E_{WtE}	331	285	275	285	454

Avoided emissions (electricity balance)

$E_{electricity}$ [a]	**Landfill**	73
	GC/SRC	165
	FBG/ICE	246
	FBG/ORC	107
	GG/SRC	131

[a] Calculated for each WtE configuration considering the electricity balance (see Table 1). A positive electricity balance means a net production of electricity.

Figure 8 shows the differential GHG impact, the GHG reduction for the replacement of the landfill by the four WtE schemes proposed. As expected all WtE configurations involve GHG reduction, concluding that the replacement of the current landfill-based system by a WtE scheme will benefit the climate impact. The use of gasification in a FBG with an ICE is shown to provide the highest climate benefit between the options considered. The low efficiency in the ORC cycle configuration involves to achieve the lowest avoided emissions being the differential GHG impact even lower than WtE scheme based on incineration.

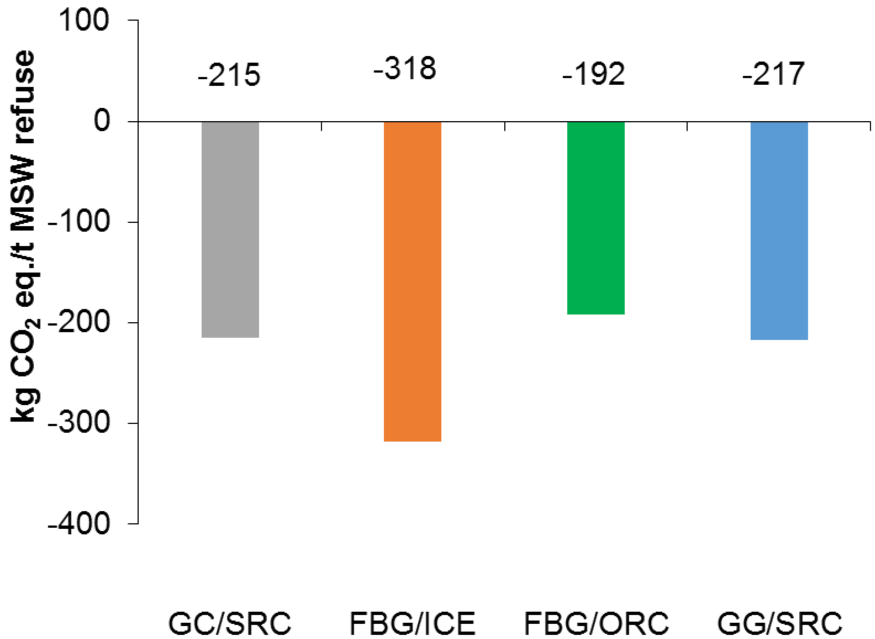


Figure 8. Differential GHG impact for the replacement of the landfill disposal for the WtE systems proposed in this study.

3.3. Selection of number WtE units in Andalusia

In Andalusia, the possible minimum and maximum plant capacities for gasification considering the production of MSW refuse in each MBT plant are 5.2 and 58.5 MWth. The average of the plant sizes for the 23 MBT plants is 20.7 MWth; However if the highest and lowest MBT plants were excluded (those with the highest standard deviations), the average would be 14.4 MWth. Figure 9 gives the influence of the gate fee on the feasibility of gasification plants for these four inputs (5.2, 14.4, 20.7 and 58.5 MWth). Only positive results are shown, in the case of minimum capacity plant, positive results are not found for the GG/SRC configuration.Since the gate fee in Southern Europe is highly dependent on the region and the waste collection system (CEWEP, 2016), a range of 20-80 €/t MSW was considered. The feasibility of the gasification WtE schemes is highly dependent on the gate fee. It is seen that for the current gate fee in Andalusia (20 €/t) a maximum of IRR of 13% would be achieved even for the maximum capacity plant (58.5 MWth) and the most efficient technology (FBG/ICE). However, with a gate fee of 60 €/t, the IRR would increase to 17% (for the same technology) for the average capacity (20.7 MWth). The figure also confirms that ICE has the highest sensitivity (higher slop) among the technologies analyzed. Overall, it seems clear that the feasibility of the replacement of the landfill by WtE schemes in Andalusia is conditioned to the increase of the gate fee, and an increase of a factor of 3 is required to envision a market penetration from the economic point of view.

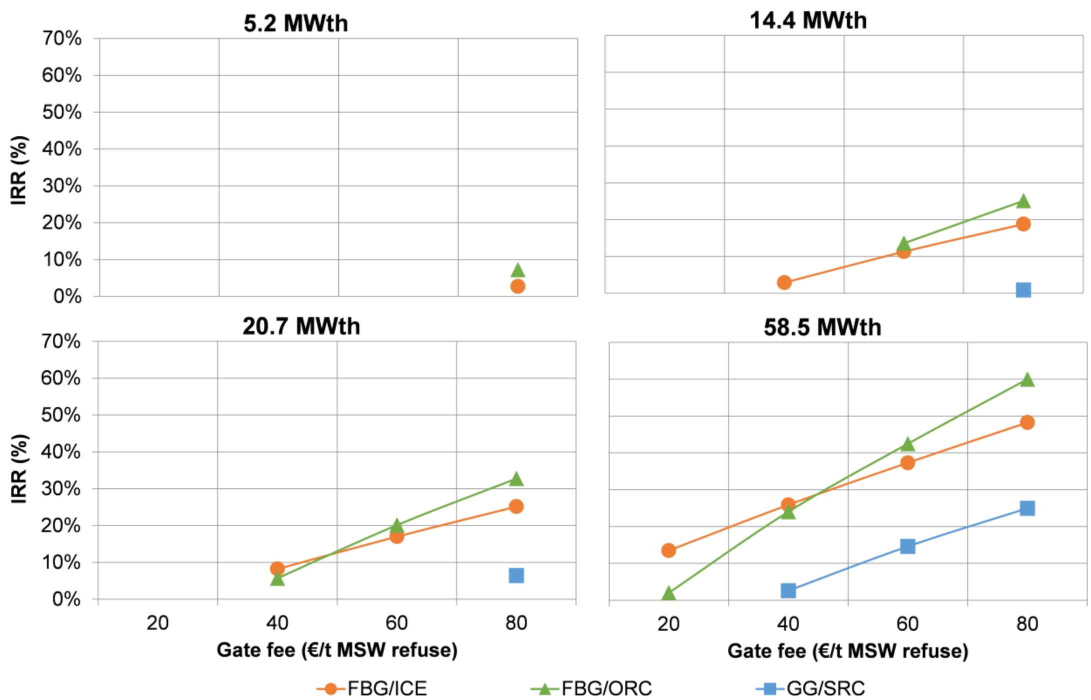


Figure 9. Sensitivity analysis to the IRR according to the current and expected gate fee for an input of 5.2, 14.4, 20.7 and 58.5 MWth.

Figure 10 shows the optimal distribution calculated considering a minimum IRR of 15%. The figure includes the location of the plants (dots for gasification and stars for incineration) with indication of the electricity output for the best gasification technologies (FBG with ORC and with ICE) and for the incineration plants. As seen incineration two centralized plants with size of 49 (East) and 48 (West) MW_e are to be set up in Andalusia. In the case of the gasification: twenty-three (one per MBT plant) FBG/ORC gasification plants could be set in Andalusia with plant capacities between 0.2 and 6.0 MW_e or FBG/ICE gasification plants with plant capacities between 0.5 and 18 MW_e.

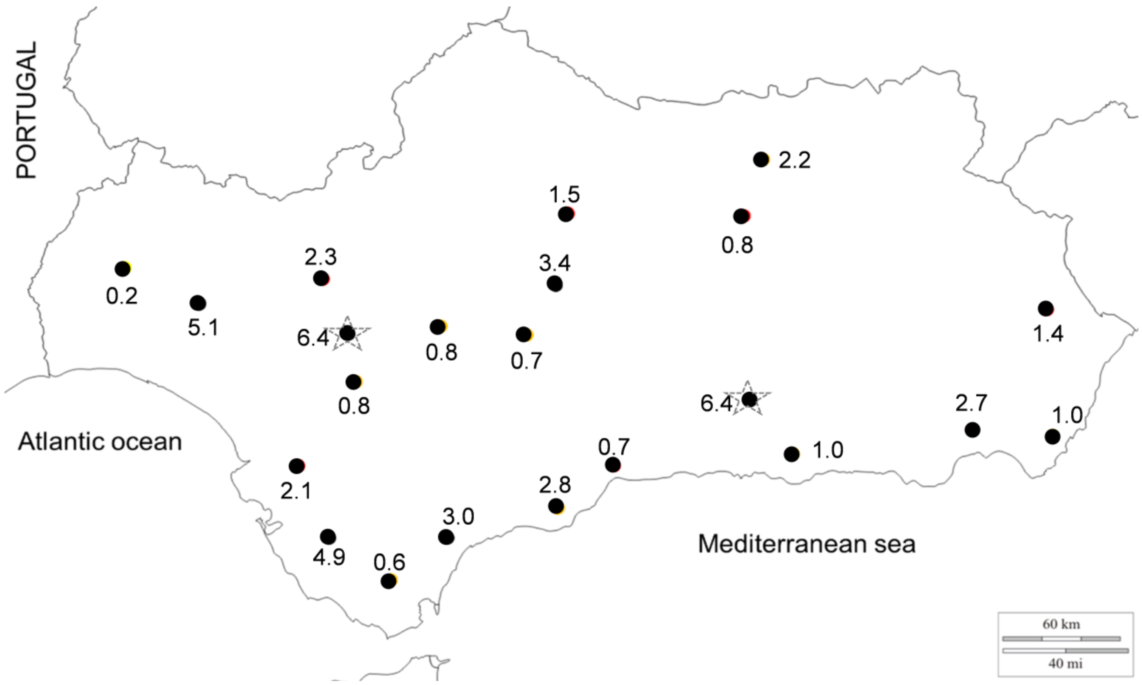


Figure 10. Location of MBT plants in Andalusia (dots). Every gasification plant is associated with a MBT plant able to produce equivalent RDF to at least 0.2 MW_e. The numbers indicate the capacity of the FBG/ORC gasification plant (MW_e) (the conversion factor for the FBG with ICE is 2.16). The highest-capacity MBT plants (stars) are selected for the location of the incineration plants (49 and 48 MWe in the East and West respectively).

Currently, the gate fee in Andalusia is very low, about 20 €/t, which explains that the MSW refuse still goes to landfill disposal. The assumed minimum gate fee to come up with feasible WtE plants is still very far (three times higher) from the current gate fee, which explains the difficulty of replacement of landfill by a WtE scheme in Andalusia in the short term, in spite of the high environmental benefit.

3.4. Sensitivity analysis

In the study, some variables were identified to have a large influence on the results presented. For the economic assessment, the parameters are the transportation costs (only considered in the incineration scheme), wholesale electricity tariff and labor costs. For the environmental assessment, the ratio of biogas recovered in the landfill is the main parameter affecting the GHG emissions.

Figure 11 shows the IRR results by modification of transport costs, wholesale electric tariff and labor costs. Wholesale electric tariff is the most influential parameter in the economic results for all the WtE schemes analyzed, especially for incineration. An increase of 20% in this tariff involves an increase of almost 29% in the IRR for incineration and between 17 and 21% for schemes based on gasification.

The three parameters analyzed change from region to region (EEA reports, 2013). Consideration of the variation of these parameters with respect to the base case analyzed here (Andalusia) can provide preliminary trends for other European regions with similar waste management scheme (landfill-dominated). Taking this in mind, and assuming other variables are similar, the results suggest that setting up WtE plants in regions with high incentive tariff could be more easily implemented, for instance in Italy with a tariff of 88.74 €/MWhe (Arena et al., 2015). Clearly, this is just a rough indication since labor and transport costs in Italy are higher than those assumed for Andalusia and the gate fee in Italy varies from about 80 €/t to about 120 €/t (AER, 2010; CEWEP, 2016). To achieve more accurate estimations, all the variables have to be taken into account at once. By doing so, the economic model developed in this work with the Italian parameters shows that WtE plants are profitable in that country, especially in regions with the highest gate fee. Considering Romania, at a first glance the most advantageous WtE scenario is incineration because of the low transportation cost in that country, but the gate fee in this country is 27 €/t since 2016 (CEWEP, 2016). Therefore, it is concluded from the sensitivity analysis made that the gate fee in Romania has to be higher before the implementation of profitable schemes based on incineration can be made.

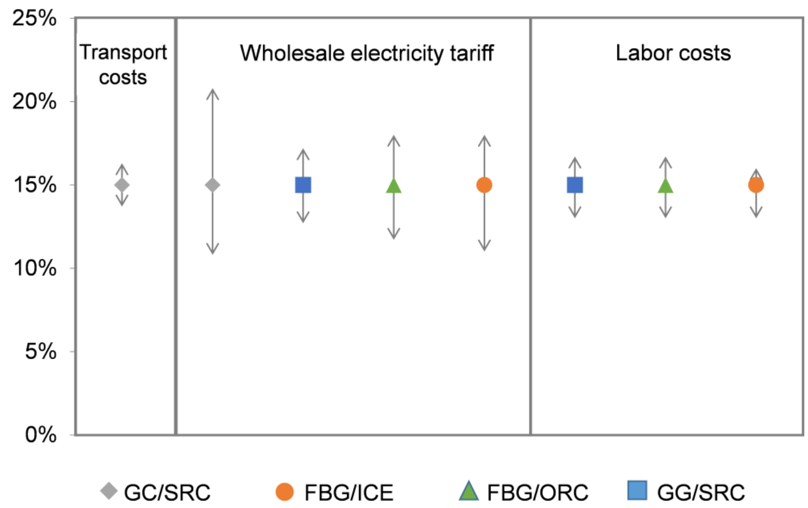


Figure 11. Sensitivity analysis of the IRR to the influential parameters in the economic assessment considering a ±20% of variability.

Figure 12 shows the sensitivity of the differential GHG impact for an interval from 0% to 95% of biogas collection. The differential GHG impact varies from ±70% in the case of the FBG/ICE to ±84% in the case of FBG/ORC. However, all configurations achieve a climate benefit as far as 95% of the biogas is collected in the landfill site.

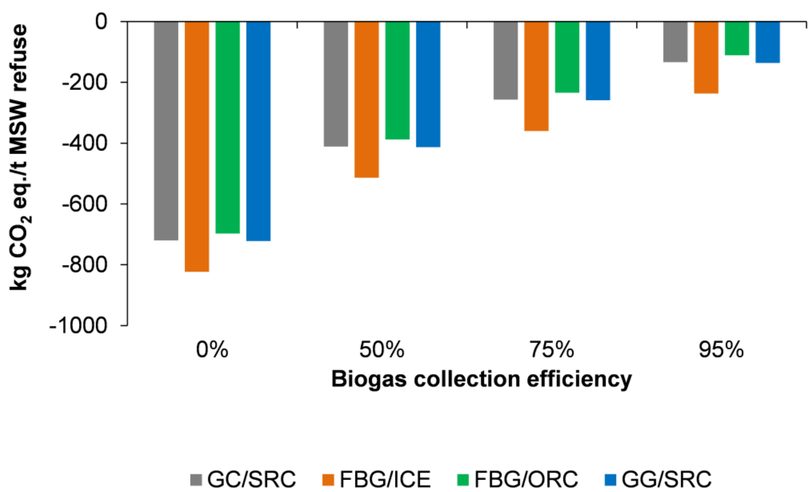


Figure 12. Sensitivity analysis of the differential GHG impact to the ratio of biogas collected in landfill site.

3.5. Discussion

In the present study, the GHG emissions associated to gasification technology are even lower than those to incineration. Therefore, the replacement of the landfill for a WtE system could result in an environmental benefit in terms of GHG emissions. *Cailean and Teodosiu* (2016) published an assessment of the Romanian solid waste management system based on sustainable development indicators. Table 9 shows some of the indicators for Romania and they are compared with Andalusian and Serbian indicators (Stanisavljevic et al., 2012). Although Andalusia has a much higher GDP (+70% regarding Romania and +80% regarding Serbia) and MSW generation ratio (+30% regarding Romania and +20% regarding Serbia) indicating Andalusia is a more developed region than those Southern European countries, the MSW ratio to landfilling is similar (-18% regarding Romania and -21% regarding Serbia) and very far of the European average (30%). Moreover, the carbon footprint associated to landfilling is also similar (-18% regarding Romania and -28% regarding Serbia).

Table 9. Comparison between Andalusia and Southern European countries (Romania and Serbia).

	Andalusia (Spain) (Eurostat)	**Romania** (Cailean and Teodosiu, 2016)	**Serbia** (Stanisavljevic et al., 2012)
GDP (€/capita)	17,263 (2016)	4,800 (2013)	4,000 (2010)
MSW generation (kg MSW/capita)	375 (2016)	272 (2013)	317 (2010)
Waste to landfilling (%)	79	96	≈100
Landfill carbon footprint (t CO_2 eq./t MSW)	0.45	0.55	0.63

In the sensitivity analysis, wholesale electricity tariff was found to have the larger impact to the IRR. Considering the transport cost in Southern European countries, results achieved in Andalusia for the incineration plants could be applicable in countries like Cyprus, Croatia and Romania (15, 10 and 5 € per hour and worker, respectively (Eurostat, 2015)). However, the gate fee for landfilling should be increased in Romania (currently 11-27 €/t (CEWEP, 2016)) and implemented in Croatia and Cyprus. In the case of the gasification schemes and considering both labor costs and wholesale electricity tariff, results achieved in Andalusia could be applicable in countries like Portugal and Greece but an increase in the gate fee would be necessary in both countries (CEWEP, 2016). From the differential GHG impact point of view, the GHG reduction would be achieved in all the European countries selected regardless the ratio of biogas collected in the landfill site. Moreover, considering the landfilling ratios and/or the content of organic matter in the MSW of these countries (EEA reports, 2013), similar GHG reduction can be expected in countries like Cyprus, Greece, Croatia and Malta.

4. Summary and conclusions

This study assesses the benefits of implementing gasification technologies in countries with dominant landfilling in Europe and where electricity is the main target from the thermal valorization point of view (i.e. low heat demand), i.e., Southern European countries. Various WtE schemes have been considered, from incineration to different options based on gasification. The study includes both techno-economic and GHG impact and is mainly focused on Andalusia, a region located in the south of Spain where 80% of the MSW generated in MBT plants are landfilled. The results are extended to consider other Southern European landfill-dominated countries (Italy, Croatia, Romania, Portugal and Greece) by applying a sensitivity analysis. The main conclusions are:

- The replacement of landfill disposal by WtE schemes in landfill-dominated European countries is positive from the environmental point of view for all WtE options analyzed, since they provide GHG reduction potential compared to landfilling
- Gasification-based compare favorably with incineration-based WtE plants. Among gasification-based WtE plants, the combined use of ORC and ICE gives the higher profitability for a given gate fee. ICE is by far the best option if the power obtained from 1 ton of refuse is also considered, due to much higher electrical efficiency. However ORC seems to be a better option in the short-term due to its higher technical reliability.

- In the case study of Andalusia, the gasification-based WtE plants have been designed as integrated with the existing MBT plants (ranging from 1.6 to 58.5 MW_{th}). For incineration-based WtE plants, an optimal allocation has been obtained resulting in two centralized WtE plants with power outputs of 49 and 48 MW_e.
- The gate fee is the main factor affecting profitability in WtE schemes. Calculations indicate that in Andalusia the gate fee has to be triple to make projects of gasification-based WtE plants profitable (IRR 15%). Otherwise, landfilling will dominate the MSW disposal in the next years despite the environmental advantages of the WtE schemes discussed.
- Considering the differences along Southern European countries, gasification plants would be economically favored in Portugal, Greece and Andalusia (Southern Spain).
- Southern European countries, regardless their economic development, have comparable GHG impact results thanks to their similar landfill carbon footprint.

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List of symbols

e_p emissions from processing (kg CO_2 eq./t MSW)

e_t emissions from feedstock transport (kg CO_2 eq./t MSW)

e_{ash} emissions from ash treatment (kg CO_2 eq./t MSW)

y_1 specific electricity production from landfill biogas (MWh_e/t MSW refuse)

y_2 specific electricity production from incineration (MWh_e/t MSW refuse)

y_3 specific electricity production from gasification (MWh_e/t MSW refuse)

Greek letters

β Biogenic fraction in the feedstock

Abbreviations

DOC Degradable organic carbon

E Global GHG emissions (kg CO_2 eq./t MSW)

$E_{landfill}$ Global GHG emissions in the landfilling (kg CO_2 eq./t MSW)

E_{WtE} Global GHG emissions in the waste-to-energy systems (kg CO_2 eq./t MSW)

EF Fossil reference (t CO_2 eq./MWh)

EPA U.S. Environmental Protection Agency

FBG Fluidized bed gasifier

FBG/ICE Fluidized bed gasifier with internal combustion engine

FBG/ORC Fluidized bed gasifier with organic Rankine cycle

GDP Gross domestic product (€/capita)

GG Grate gasifier

GC Grate combustor

GC/SRC Grate combustor with steam Rankine cycle

GG/SRC Grate gasifier with steam Rankine cycle

GHG Greenhouse gases

ICE Internal combustion engine

IPCC Intergovernmental panel on climate change

IRR Internal return rate

LCA Life cycle assessment

LHV Lower heating value

MBT Mechanical and biological treatment

MSW Municipal solid waste

ORC Organic Rankine cycle

RDF Refuse derived fuel

RED Renewable energy directive

SRC Steam Rankine cycle

TIC Total investment costs

TOC Total operational costs

USA United States of America

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Paper VI

Dynamic assessment of Waste-to-Energy schemes in current European landfill-dominated regions

In this paper, advanced Waste-to-Energy (WtE) schemes based on gasification are proposed in order to minimize the landfill disposal in European landfill-dominant regions. Technical development of advanced WtE plants (gasification-based), evolution of waste management schemes according to realistic European targets and electricity production mix, as well as the environmental impact to a changing European society are considered in the paper. It has been accepted as conference proceedings in 2017[17].

1. Introduction

Even when European regulation has been encouraging landfill reduction in the last decades (Directive 2008/98/EC), 13 out of 28 EU countries still landfill more than 50% of their municipal solid waste (MSW), mainly located in Southern and Eastern Europe (Figure 1). Landfill disposal requires the use of a not always available land and has several environmental impacts associated (land, atmosphere, hydrosphere and biosphere). In fact, recent studies determine the methane concentration in the atmosphere has dramatically rised in last decades. This methane is released from different sources but two thirds of the emissions are attributable to anthropogenic activities related to agriculture and waste management (Saunois et al., 2016, Global Carbon Project, 2001). Because of that, it is necessary to find alternatives to manage the huge amount of urban wastes going to landfill disposal in Southern and Eastern Europe. MSW refuse is the unsorted stream of MSW going currently to landfill disposal or incineration. MSW refuse usually contains a biodegradable fraction over 50% (IEA, 2012). On the other hand, waste disposal (landfilling or mass-burnt incineration) should be replaced by waste-to-resource alternatives in order to reduce GHG emissions. In this study, advanced Waste-to-Energy

[17] Aracil C, Pedro Haro P, Fuentes-Cano D, Gómez-Barea A. Dynamic assessment of Waste-to-Energy schemes in current European landfill-dominant regions. EUBCE 2017.

(WtE) schemes based on gasification are proposed in order to minimize the landfill disposal in European landfill-dominated regions. Technical development of advanced WtE plants, evolution of waste management schemes according to realistic European targets and electricity production mix, as well as the environmental impact to a changing European society are considered in the study. In order to do so, a previously developed dynamic GHG emission assessment methodology is used (Aracil et al. 2017). A dynamic assessment is crucial when comparing with a dynamic reference system (i.e. methane emissions are delayed several months or years after the landfilling of the wastes and the emissions continuing for at least 20 years more). Furthermore, the evolution of current waste management and electricity production needs to be modeled and is, therefore, included in the study.

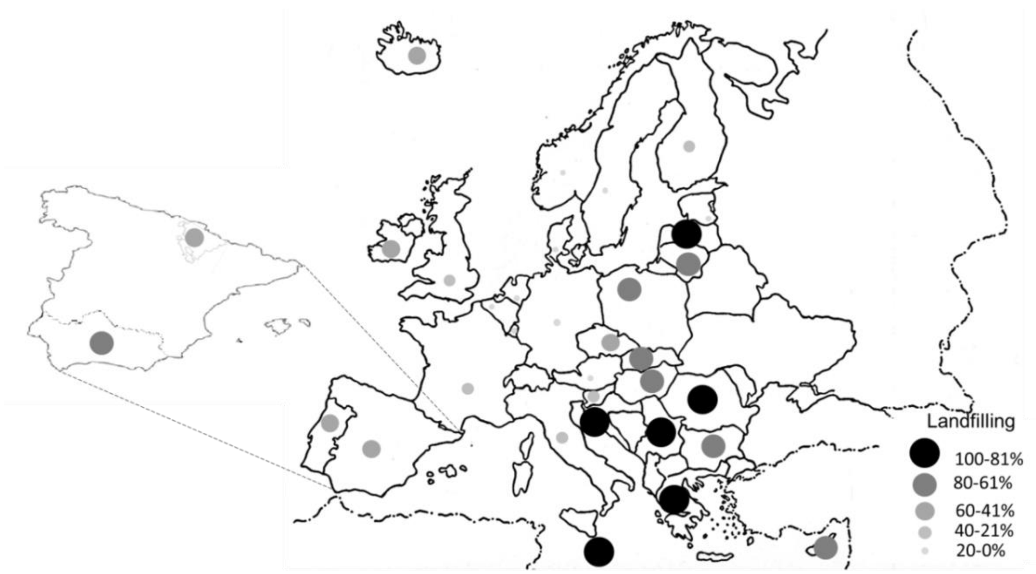

Figure 1. Landfilling ratios of MSW refuse in each European country (Eurostat, 2013) and specifically in two Spanish regions (South: Andalusia, North: The Basque Country).

Up to now, only the identification of the potential energy recovery and discussion of the technical and economic feasibility of advanced WtE schemes had been developed. The results indicate that the production of electricity is a feasible option at short term and that the impact of using MSW refuse as feedstock is better than reported in the scarce existing literature. However, the economic results are strongly dependent on the gate fee and wholesale electric tariff for each country. The environmental aspects have not been fully discussed considering the consequential impact. Up to our previous study (Aracil et al. 2017), a dynamic assessment has not been made.

2. Goal and scope

The aim of this study is the dynamic GHG emission assessment of advanced WtE schemes based on gasification ir order to replace the landfill disposal. Three diferent configurations of advanced WtE plants according to the type of reactor and form of electricity production are proposed: grate gasifier with steam Rankine cycle (GG/SRC), fluidised bed gasifier with organic Rankine cycle (FBG/ORC) and FBG with internal combustion engine (FBG/ICE). The functional input unit is 1 ton of MSW refuse and the output is electricity. Figure 2 shows a simplified diagram of the WtE schemes proposed.

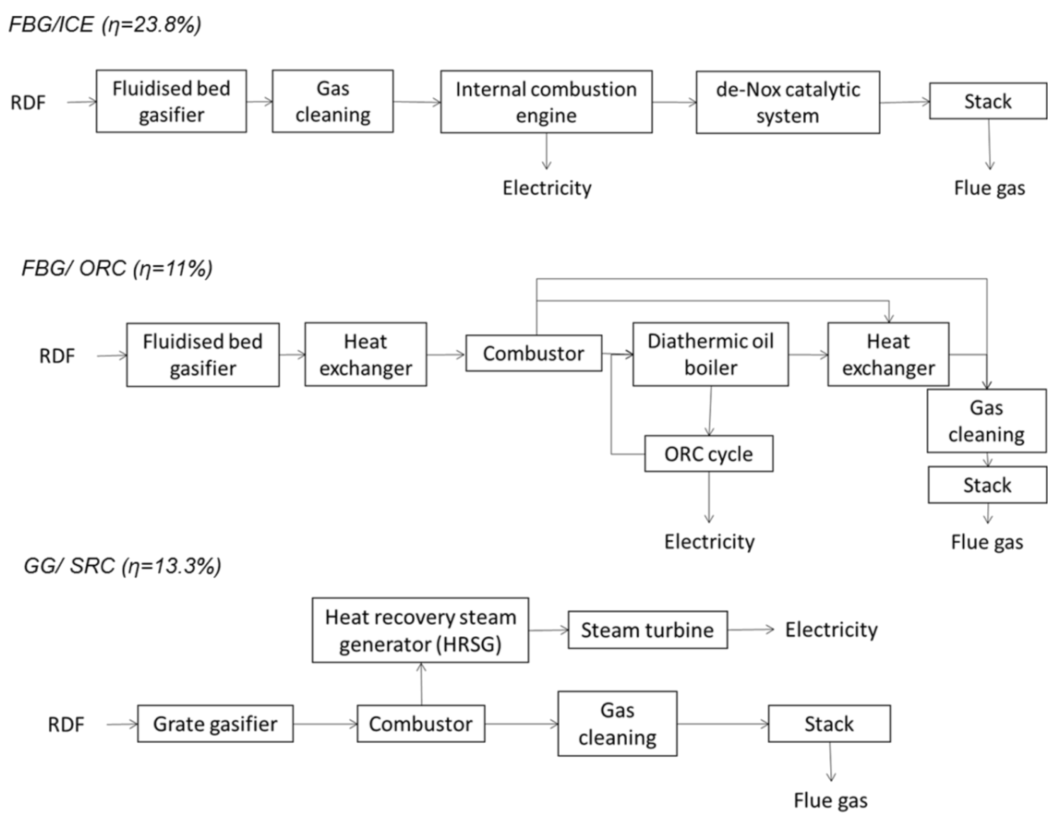

Figure 2. Simplified diagrams of the WtE based on gasification plants proposed according to the gasification technology and the electricity production system.

3. Methodology

The climate benefit indicator chosen for this study is the climate mitigation index (CMI) (Aracil et al., 2017) which compares the behavior of the WtE plant with the current MSW management, and production of products and services for a specific region (Table 1).

Table 1. Summary of the main characteristics of the CMI

	Climate Mitigation Index (CMI)
Based on	AGWP (cumulative)
Units	--
Emissions included	Biogenic and anthropogenic emissions
Comparison	BIO and BAU system for the same region
Result	Cumulative climate mitigation for a specific region

Two different scenarios are taken into account: Scenario 1, in which the reference system (landfill) remains unaltered, and Scenario 2, in which there is an evolution towards landfill banning and decarbonization of the energy mix (Figure 3 and 4).

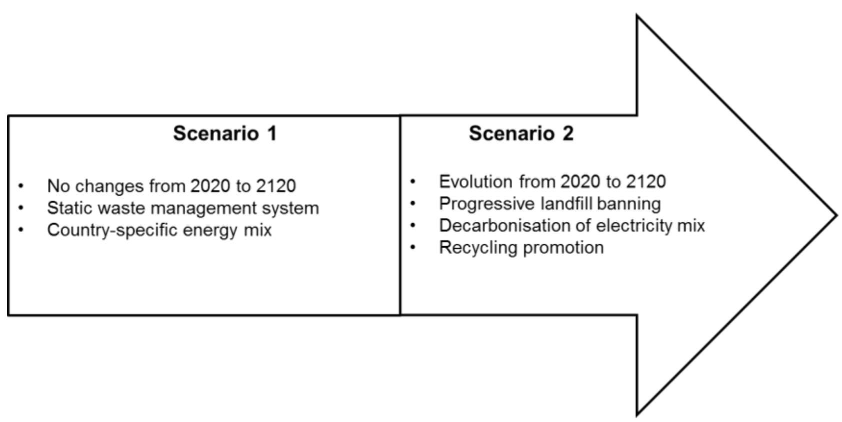

Figure 3. Proposed scenarios in this study

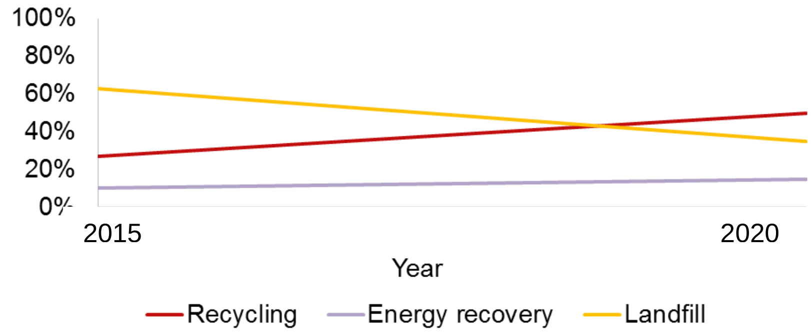

Figure 4. The evolution of waste management schemes in Scenario 2.

The case study for this study is the region of Andalusia in Southern Spain. The Spanish waste management scheme is based on recycling and landfilling and, to a lesser extent, on incineration. However, in Andalusia landfilling is dominant and incineration is not implemented, as commonly found in Southern and Eastern European countries.

4. Results

Figure 5 shows the CMI of the three advanced WtE plants (based on gasification) proposed in this study in Scenario 1 (a) and Scenario 2 (b). In scenario 1, there is a sharp reduction of the index from positive to negative between years 3 to 8 depending on the plant configuration achieving the best results the FBG with ICE configuration and the worst the FBG with ORC configuration. Then the trends increase towards the climate worsening (from negative to positive) between years 65 and 80 for FBG/ORC and GG/SRC options. The climate mitigation index in the FBG/ICE option is negative from year 8. The results are according to the energy efficiency. The highest efficiency, the highest climate mitigation. In scenario 2, the period of climate mitigation is shortened and all the options, including FBG/ICE, achieve the climate worsening in the last years when the climate impact from the incenration is avoided. In all cases, the highest climate mitigation is achieved at a short time (first 20 years) since the transient emissions from the landfill are concentrated around 20 years after the landfilling of the MSW refuse.

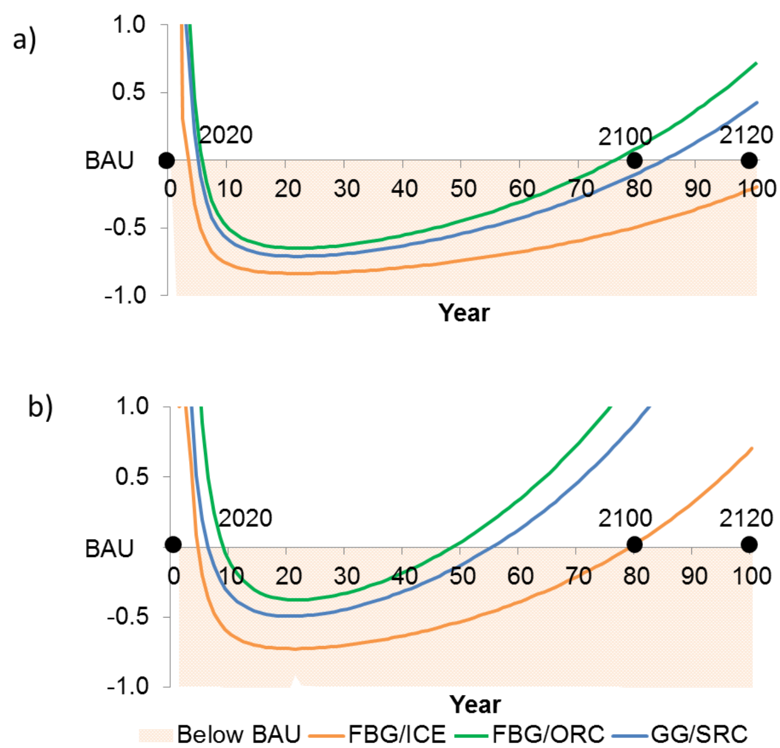

Figure 5. Climate mitigation index (CMI) of the three advanced WtE plants proposed based on gasification in Scenario 1 (a) and Scenario 2 (b)

5. Conclusions

The results reveal that the incorporation of gasification-based WtE plants in dominated-landfill European countries has a positive climate impact compared to current waste management in the short term. The long-term climate impact is, however, not secure since it depends on the evolution of the reference system in the analyzed region. In fact, if the evolution towards energy recovery (incineration) in a landfill-dominated country is considered (scenario 2), a climate worsening is achieved for all the gasification-based configurations at a long term. Among the assessed configurations, the FBG/ICE configuration achieves the best climate benefit since has the highest energy efficiency.

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Curriculum vitae

Cristina Aracil was born in Seville in 1983. From 2001 to 2006, she studied Environmental Sciences at the Pablo de Olavide's University in Seville (long-cycle, 5 years, degree). From 2008 to 2010, she worked as Environmental Inspector in the Environmental Management Company (EGMASA, public company). From 2010 to 2012, she studied a Master on Renewable Resources and Energy Engineering at the University of Extremadura (Badajoz, Spain). In 2012, she joined the Bioenergy Group (BEGUS) at the University of Seville as PhD student. In 2016, she lectured Environmental Engineering at the University of Seville.

Cristina's main research areas are waste management, modeling of static and dynamic GHG emissions assessments and modeling of greenhouse gas removal (GGR) technologies.

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