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Evaluation du potentiel d'émissions négatives des technologies d'utilisation du CO2

Présentée par Sibylle Duval--Dachary
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Sous la direction de Arnaud HELIAS

Devant le jury composé de

Annie LEVASSEUR, Professeure agrégée, Université du Québec	Rapporteure
Philippe LOUBET, Maitre de conférences, Bordeaux INP	Rapporteur
Cécile BESSOU, Directrice de recherche, CIRAD	Examinatrice
Benoît GABRIELLE, Professeur, AgroParisTech	Examinateur
Sandrine SELOSSE, Chargée de recherche, Mines Paris - PSL	Examinatrice
Pierre COLLET, Docteur, Quantis	Invité
Arnaud HELIAS, Directeur de recherche, INRAE	Directeur de thèse

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Guillaume Batôt
Sandra Beauchet
Daphné Lorne

UMR - ITAP

Arnaud Hélias
Amandine Pastor
Thibault Salou

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Abstract

The Intergovernmental Panel on Climate Change raised awareness on the urgent need for action to limit climate change. Answers include Carbon Capture and Utilization (CCU) and Negative Emissions Technologies (NET). Life Cycle Assessment (LCA) studies of CCU systems are increasingly being carried out. Since 2020, three guidelines have been published in order to harmonise them. CCU systems can be designed to produce negative emissions (CCUNET). However, the coupling with negative emission technologies raises issues regarding the definition of the system boundaries, the timing of uptake and release of atmospheric CO_2, the choice of the reference system, and the solving of multifunctionality. This brings us to the research question of this thesis: How to address the methodological challenges associated with the LCA of CCUNET systems? Firstly, bioenergy with carbon capture and storage is identified as the most probable source of atmospheric CO_2 removal in the near future. A literature review is performed to compile generic inventory tables for facilitating the use of "from-cradle-to-grave" system boundaries. Secondly, the compatibility of LCA guidelines for CCU systems with the evaluation of negative emissions is assessed, resulting in recommendations on the definition of the functional unit and the methods to account for atmospheric CO_2 and to deal with multifunctionality. Thirdly, dynamic LCA is the appropriate answer to account for differences in the dynamics of carbon storage by photosynthesis and releases at the end-of-life. How to make dynamic LCA easier to use is thus explored. Three purposes are addressed, the modelling tools, the time dimension in the functional unit and the contribution of the time dimension to the accuracy of results. Dynamic LCA demands more data on the system and increases the complexity of the calculation, but the precision of the results is not necessarily significantly increased. Lastly, a method is thus proposed to determine if the dynamic approach will significantly change the results compared to a static approach with only a little knowledge of the dynamics of the system under study.

Résumé

Le groupe d'experts intergouvernemental sur l'évolution du climat alerte sur l'urgence d'agir pour limiter le changement climatique. Le captage et l'utilisation du carbone (CCU) ainsi que les technologies à émissions négatives (NET) font partie de la solution. Les analyses de cycle de vie (ACV) sur les systèmes CCU se multiplient. Depuis 2020, trois guides méthodologiques ont été publiées afin de les harmoniser. Les systèmes CCU peuvent être conçus pour produire des émissions négatives (CCUNET). Cependant, ce couplage soulève des questions sur la définition des limites du système, sur les différences temporelles entre l'absorption et la libération du CO_2 atmosphérique, sur le choix du système de référence et sur la résolution de la multifonctionnalité. Cela nous amène à la question de recherche de cette thèse : comment relever les défis méthodologiques associés à l'ACV des systèmes CCUNET ? Tout d'abord, la bioénergie avec capture et stockage du carbone est identifiée comme la source la plus probable de fixation du CO_2 atmosphérique dans un avenir proche. Une revue de la littérature est réalisée pour compiler des tables d'inventaire génériques afin de faciliter l'utilisation des limites des systèmes "du berceau à la tombe". Deuxièmement, la compatibilité des guides méthodologiques pour l’ACV de systèmes CCU avec l'évaluation des émissions négatives est évaluée, donnant lieu à des recommandations sur la définition de l'unité fonctionnelle et les méthodes de prise en compte du CO_2 atmosphérique et de traitement de la multifonctionnalité. Troisièmement, l'ACV dynamique est la réponse appropriée pour prendre en compte la dimension temporelle des émissions de CO_2 atmosphérique. La facilitation de l’usage de l'ACV dynamique est explorée à travers trois objectifs : les outils de modélisation, la dimension temporelle dans l'unité fonctionnelle et la contribution de la dimension temporelle à la précision des résultats. L'ACV dynamique demande plus de données sur le système et augmente la complexité du calcul, mais la précision des résultats n'est pas nécessairement augmentée de manière significative. Une méthode est donc proposée en dernière partie pour déterminer si l'approche dynamique modifiera significativement les résultats par rapport à une approche statique, avec seulement une faible connaissance de la dynamique du système étudié.

Résumé étendu

CONTEXTE, QUESTION ET STRATEGIE DE RECHERCHE

Les activités humaines depuis la révolution industrielle sont à l'origine d'une hausse de la concentration de CO_2 dans l'atmosphère de 130 ppm. Cela représente près de 80% des émissions anthropiques de gaz à effet de serre (GES), les 20% restant étant principalement liés aux émissions de méthane et de protoxyde d'azote. Le CO_2 est ainsi le principal contributeur à la hausse du forçage radiatif dans le monde au cours du 20^{e} siècle, avec déjà une augmentation de 1°C de la température mondiale (Introduction, section 1).

Pour limiter l'augmentation future à 2°C voire 1.5°C, de plus en plus de pays et d'entreprises se fixent des objectifs de neutralité carbone à horizon 2050-2060. Cependant, au rythme actuel de la baisse des émissions de GES, les scénarios à 1.5°C voire 2°C semblent difficilement atteignables. C'est pourquoi l'IPCC dans son 5^{e} rapport a souligné l'importance du développement des technologies à émissions négatives (NET). Ces NET capturent volontairement du CO_2 de l'atmosphère pour le stocker de manière permanente.

Les NET regroupent des procédés biologiques, comme la séquestration du carbone dans le sol, et des procédés industriels, comme la minéralisation du CO_2. Les stratégies de l'Europe et de la France (Introduction, section 2) incluent ce type de technologies, ainsi que des technologies de capture et utilisation du CO_2 (CCU). Le CCU consiste à valoriser le CO_2 capturé, avec transformation chimique par exemple en carburant ou encore en matière plastique, ou sans transformation chimique par exemple pour une utilisation en tant que solvant. Le CCU est notamment une solution pour décarboner l'industrie chimique qui utilise encore majoritairement des matières premières fossiles. Coupler NET et CCU est donc une opportunité de générer à la fois des émissions négatives, des produits de valeur et d'éviter l'utilisation de matières premières fossiles (Introduction, section 3). La pertinence environnementale de ces systèmes, nommés CCUNET, doit être évaluée à l'aide de l'analyse de cycle de vie (ACV).

L'ACV est la méthodologie normée pour l'évaluation environnementale de biens et services. Elle se déroule en quatre étapes, avec des itérations si besoin. Elle commence par la définition des objectifs et du cadre de l'étude. À cette étape, les questions auxquelles l'ACV répondra sont définies. Par exemple : "Le produit A est-il environnementalement meilleur que le produit B ?". Ensuite, les choix méthodologiques et les principales hypothèses sont explicitement formulés pour définir le cadre dans lequel les résultats de l'ACV sont valides. Cela inclut notamment les frontières du système étudié, l'unité fonctionnelle (UF) ou encore la méthode pour résoudre la multifonctionnalité si nécessaire (si le système a plusieurs produits). Ensuite des données sont collectées pour calculer l'inventaire du cycle de vie du système. Un inventaire rassemble toutes les émissions vers l'environnement ainsi que la consommation de ressources causées par le système étudié. La collecte de données pour construire l'inventaire est une partie essentielle (et chronophage) de la réalisation d'une ACV. Les données proviennent de diverses sources (modélisation du système, mesures sur le terrain, littérature, bases de données). Puis l'inventaire est converti en impacts potentiels sur l'environnement grâce à des facteurs de caractérisation. Au cours de cette étape d'évaluation de l'impact du cycle de vie, plusieurs catégories d'impacts sont considérées (changement climatique, acidification, eutrophisation...) pour pouvoir identifier les éventuels compromis entre les catégories. Enfin, les contributions des substances et des processus à l'impact sont analysées pour identifier les erreurs et juger de la pertinence des hypothèses de modélisation (étape d'interprétation). Par exemple, si un processus contribue de manière significative à l'impact, le praticien de l'ACV peut décider d'améliorer l'inventaire pour ce processus en particulier.

L'ACV est une méthode mature qui soulève encore des questions méthodologiques, autour de la définition des frontières du système, de la comptabilisation du carbone atmosphérique ou encore de la prise en compte de la dimension temporelle (Introduction, section 4). D'où la question de recherche de cette thèse : comment aborder les défis méthodologiques associés à l'ACV des systèmes CCUNET ? La stratégie de recherche est la suivante (Introduction, section 5) :

- Chapitre 1 : Fournir des données d'inventaire pour pouvoir utiliser des frontières du système « du berceau à la tombe ». Pour évaluer le potentiel d'émissions négatives, toutes les étapes du cycle de vie doivent être incluses dans les limites du système, de la capture du CO_2 dans l'atmosphère à son stockage permanent hors de l'atmosphère ou sa libération.
- Chapitre 2: Explorer la compatibilité des guides méthodologiques pour l'ACV des systèmes CCU avec l'évaluation des émissions négatives.
- Chapitre 3 : Faciliter l'ACV dynamique pour pouvoir inclure la dimension temporelle dans l'évaluation de l'impact sur le changement climatique, notamment pour la capture et la réémission du CO_2 atmosphérique.
- Chapitre 4 : Proposer une méthode pour évaluer si l'ACV dynamique est nécessaire en utilisant uniquement des informations temporelles simplifiées pour permettre aux praticiens de l'ACV de cibler leurs efforts de manière plus efficace entre l'amélioration de la qualité des données d'inventaire et la réalisation de l'ACV dynamique.

CHAPITRE 1: ANALYSE DU CYCLE DE VIE DE LA BIOENERGIE AVEC SYSTEME DE CAPTURE ET DE STOCKAGE DU CARBONE : REVUE CRITIQUE DES INVENTAIRES DE CYCLE DE VIE

Ce premier chapitre est lié à l'utilisation des frontières du système de type « du berceau à la tombe » pour réaliser l'évaluation des émissions négatives. Pour faciliter leur utilisation, des données d'inventaire doivent être disponibles pour les étapes du cycle de vie qui ne sont pas le principal sujet de l'étude ACV. Pour les systèmes CCUNET, il s'agira probablement de la production en amont de CO_2 (source de CO_2) et des étapes de fin de vie. Les systèmes de bioénergie avec capture et stockage de CO_2 (BECCS) sont les sources les plus probables de CO_2 atmosphérique (i.e. qui provient de l'atmosphère) dans un avenir proche (Chapitre 1, section « Chapter context »). La qualité des données d'inventaire est essentielle pour la fiabilité des résultats de l'ACV. Cela conduit à la question explorée dans le premier article : quelles données d'inventaire sont utilisées pour l'évaluation du cycle de vie des BECCS ? Ce chapitre est un article publié : Duval-Dachary S, Beauchet S, Lorne D, Salou T, Helias A, Pastor A (2023) Life cycle assessment of bioenergy with carbon capture and storage systems: Critical review of life cycle inventories. Renewable and Sustainable Energy Reviews 183:113415. https://doi.org/10.1016/j.rser.2023.113415.

Matériel et méthode (Chapitre 1, section 2)

Pour réaliser cette revue, les bases de données 'Scopus' et 'Web of science' ont été explorées à l'aide d'une requête combinant des mots clés décrivant les concepts de 'bioénergie', 'ACV' et 'stockage de carbone'. Un ensemble de 97 articles a été identifié, dont seulement 35 décrivent une ACV de systèmes BECCS. Ces 35 articles ont été analysés, pour en extraire les descriptions des systèmes étudiés (type de biomasse, de procédé de transformation, de procédé de capture de CO_2…), les données d'inventaire fournies, ainsi que des métadonnées associées (type de source, date de production, intervalle de variation…). L'étude de la qualité des données se base sur les critères précisés par la norme ISO : exhaustivité, reproductibilité, représentativité, fidélité, facteur temporel, et source des données.

Résultats et discussion

Les systèmes BECCS sont constitués de quatre grandes étapes : la production de la biomasse, sa conversion en énergie, la capture du CO_2, et enfin le transport et le stockage du CO_2. Plusieurs options existent pour chaque étape. Par exemple la biomasse peut être transformée par combustion, gazéification, fermentation, etc. Cette multiplicité d'options entraine une très grande diversité des systèmes BECCS. Dans les articles sélectionnés, les options les plus étudiées sont la combustion et la gazéification, la capture de CO_2 par solvant amine et le transport du CO_2 par pipeline. Certaines briques technologiques mériteraient d'être plus étudiées, et notamment les fermentations anaérobies, les procédés de seconde génération pour la capture du CO_2 et les options alternatives pour le transport du CO_2, telles que le train ou le camion (Chapitre 1, sections 3.1 et 4.2).

Transmettre les données d'inventaire est essentiel pour permettre la reproductibilité de l'étude. Cependant seul un tiers des articles fournit des tables d'inventaire, ce qui révèle un problème de mise à disposition des données dans la littérature. Cela peut s'expliquer lorsque les données sont protégées par le secret industriel, mais les résultats de cette revue indiquent que les données proviennent en majorité de la littérature (bases de données, articles) ou de simulations. Les données sont en effet rarement de première main, c'est-à-dire des mesures de terrain ou des données industrielles. (Chapitre 1, sections 3.2.1, 3.2.3 et 4.1.3)

Cette revue révèle également un manque de robustesse de certaines des données, anciennes ou provenant d'une seule source, comme la quantité de charbon actif nécessaire pour réaliser la capture du CO_2 par solvant amine. Les discussions sur la pertinence des données d'inventaire utilisées par rapport à l'objectif de l'ACV sont insuffisantes dans les articles sélectionnés. Des études de variabilité et d'incertitude sont réalisées, mais les autres critères, comme la cohérence géographique ou temporelle des données, ne sont pas discutés. (Chapitre 1, section 4.1.1)

Concernant l'exhaustivité des inventaires, les infrastructures ne sont pas mentionnées dans un tiers des articles. L'impact des changements de stock de carbone du sol dus à la culture de la biomasse, ainsi qu'au changement d'utilisation des terres, n'est pas pris en compte dans deux tiers des articles. De plus environ la moitié des articles calcule l'impact seulement pour la catégorie « changement climatique » et n'inclue donc pas dans leurs inventaires les émissions autres que les GES. (Chapitre 1, sections 3.2.2 et 4.1.2)

Des tables d'inventaire génériques ont tout de même pu être compilées et proposées pour la récolte et le prétraitement, la combustion et la gazéification de la biomasse, la capture par solvant amine, le transport par pipeline et le stockage géologique du CO_2. Les tables d'inventaire fournies ne doivent bien évidemment pas remplacer les données mesurées ou simulées lorsque c'est possible, mais servent de valeurs par défaut (Chapitre 1, sections 3.3 et 4.3).

Conclusion (Chapitre 1, section « Chapter conclusion »)

Cette revue de la littérature a révélé plusieurs limitations de la gestion des données dans les ACVs. La majorité des articles examinés ne discute pas l'adéquation de la qualité de leurs données d'inventaire à l'objectif de l'étude. Cette revue a également révélé un manque de disponibilité des données d'inventaire.

Cas d'étude pour cette thèse – gestion des données

Dans cette thèse, un cas d’étude est nécessaire pour illustrer les questions méthodologiques soulevées dans les chapitres 2 et 3. En ce qui concerne la transparence et la reproductibilité, toutes les données d’inventaire utilisées dans cette thèse sont fournies en tant que matériel supplémentaire des articles sous forme de fichiers Excel, ainsi que les scripts utilisés pour calculer l'ACV dans Brightway2 (librairie python pour l’ACV). Les résultats restent illustratifs et ne sont pas destinés à être utilisés pour décider si le cas d’étude doit être déployé. C'est pourquoi la qualité des données d’inventaire du cas d’étude n'est pas discutée.

L'évaluation du potentiel d'émissions négatives nécessite des frontières du système « du berceau à la tombe ». Le cas d’étude doit donc inclure une source de CO_2, un processus de valorisation du CO_2 et la fin de vie du produit à base de CO_2. La sélection du cas d’étude a été motivée par la disponibilité des données d'inventaire, par la maturité des technologies par rapport à un déploiement à 2050 et par son intérêt pour illustrer les questions méthodologiques. Une usine d'éthanol est choisie comme source de CO_2, car c’est le seul type de BECCS déjà en opération. Les biomasses sont sélectionnées pour représenter la diversité des biomasses disponibles et les défis méthodologiques associés (maïs, miscanthus et résidus de bois). En ce qui concerne la valorisation du CO_2, le méthanol est un élément essentiel de l'industrie chimique et peut être produit à partir du CO_2. La technologie de conversion du méthanol en propylène est mature et déjà utilisée à l'échelle industrielle en Chine. Un sac plastique réutilisable en polypropylène est choisi comme produit, car il s'agit d'un produit de tous les jours. Son traitement en fin de vie est de l’incinération avec capture et stockage de CO_2, ce qui permet d’utiliser les tables génériques d’inventaire compilées dans la revue et modélisant la combustion, le captage du CO_2 par le solvant amine, le transport et le stockage du CO_2.

CHAPITRE 2: ANALYSE DU CYCLE DE VIE DU CAPTAGE ET DE L'UTILISATION DU CARBONE EN TANT QUE TECHNOLOGIE D'EMISSIONS NEGATIVES : RECOMMANDATIONS ET CAS D’ETUDE

Dans le chapitre précédent, un premier enjeu méthodologique, la définition des frontières du système, a été examiné et les données collectées facilitent l’utilisation de frontières "du berceau à la tombe". Cependant, l'évaluation des émissions négatives soulève d'autres questions. La norme ISO qui encadre la pratique de l’ACV peut être appliquée à tout type de produit ou service. Certains choix méthodologiques (gestion de la multifonctionnalité, inclusion ou non de certaines étapes du cycle de vie, etc.) peuvent entraîner des divergences entre les résultats d’ACV de produits similaires. C’est pourquoi des guides méthodologiques propres à des groupes de produits, tels que les systèmes CCU ou les plastiques ont été écrits pour harmoniser les résultats et améliorer la comparabilité entre études. Malheureusement, ces guides ne contiennent pas de recommandations précises pour la réalisation d'une ACV d’un système CCUNET. Il n’existe d’ailleurs même pas de guide méthodologique exhaustif pour l’ACV des systèmes NET. Par contre de nombreux articles discutent des défis méthodologiques soulevés par l’évaluation d’émissions négatives et proposent des recommandations. Trois points sont particulièrement discutés : la temporalité de capture et de réémission du CO_2 atmosphérique, le choix du système de référence et la méthode pour gérer la multifonctionnalité. Réaliser l’ACV d’un système CCUNET va donc générer des interrogations spécifiques (Chapitre 2, section 1). Cela conduit à la question traitée dans le deuxième article : les guides méthodologiques pour l’ACV de systèmes CCU sont-ils compatibles avec l'évaluation d’émissions négatives ? Ce chapitre a été soumis à l’« International Journal of Life Cycle Assessment » le 5 février 2024 : Duval-Dachary S., Lorne D., Beauchet S., Salou T., Hélias A. *Life cycle assessment of carbon capture and utilisation as a negative emission technology: recommendations and case study*, et est actuellement en révision.

Matériel et méthode

La pertinence des recommandations doit être évaluée en lien avec les objectifs des études. Deux objectifs sont donc définis : vérifier la pertinence environnementale du système CCUNET par rapport à une situation de référence, et évaluer la quantité d’émissions négatives qu’il peut générer (propriété intrinsèque au système). La comparaison à un système de référence soulève la problématique de la définition de l’unité fonctionnelle. Les résultats sont calculés dans un premier temps sans inclure « l’élimination de CO_2 atmosphérique », fonction propre aux systèmes NET, dans l’unité fonctionnelle. Les données d’inventaire sont ensuite utilisées pour évaluer les variations qui seraient engendrées par l’ajout de cette fonction dans l’unité fonctionnelle du système CCUNET et de son système de référence (Chapitre 2, section 2.1). L’évaluation du potentiel d’émissions négatives soulève la problématique de la gestion de la multifonctionnalité. En effet, un système CCUNET a de multiples produits : ceux de la source de CO_2 et ceux produits à partir du CO_2. Un objectif de l’ACV peut être d’évaluer le potentiel d’émissions négatives du produit à base de CO_2 uniquement. Il est alors nécessaire de repartir l’impact du système complet entre ces différents produits. Trois recommandations sont comparées : l’allocation sur le contenu carbone, la substitution et la « circular footprint formula », cette dernière étant la méthode d’allocation recommandée au niveau européen. L’extension des frontières du système est également étudiée. Cette dernière signifie étendre les limites du système jusqu'à ce qu'elles englobent le cycle de vie complet de chaque coproduit. L'extension des frontières du système ne permet donc pas d’attribuer à chaque coproduit une fraction de l’impact total du système, mais permet de suivre les flux de carbone atmosphérique depuis le captage du CO_2 jusqu'à son rejet ou son stockage permanent sans distorsion. L'extension des frontières du système a donc été choisie comme méthode de référence pour évaluer le potentiel d'émissions négatives (Chapitre 2, section 2.2).

La solution pour inclure la temporalité de la capture et de la réémission du CO_2 atmosphérique est l'ACV dynamique, qui fera l'objet d'un chapitre à part entière. L'ACV dynamique ne peut être appliquée que si le CO_2 atmosphérique est comptabilisé dans l'inventaire. L'applicabilité de la méthode « +1/-1 » est donc testée : comptabiliser complètement les captures et réémissions de CO_2 atmosphérique, dans l'inventaire, et lors de la caractérisation de l'impact. Un cas d'étude est modélisé pour illustrer les recommandations et vérifier leur applicabilité. Il contient la production de biomasse (maïs), sa transformation en énergie et CO_2 (production d'éthanol), la valorisation du CO_2 en un sac en polypropylène, et enfin l'incinération, avec capture et stockage du CO_2, de ce sac (Chapitre 2, section 2.3).

Résultats et Discussion

Il n'y a pas de consensus actuellement concernant le flux de référence à utiliser pour quantifier l'unité fonctionnelle liée à la fonction d'élimination du CO_2 atmosphérique. Certains proposent d'utiliser la quantité de CO_2 stockée de manière permanente, d'autres le résultat de l'ACV dans la catégorie « changement climatique ». Les résultats montrent que le choix du flux de référence influence la définition des systèmes. Utiliser la quantité de CO_2 atmosphérique traitée permet de ne pas avoir à ajuster artificiellement les frontières des systèmes pour qu'ils répondent à la même fonction. Le traitement du CO_2 atmosphérique fait référence à tout captage de CO_2 atmosphérique (dans l'atmosphère ou provenant d'une industrie basée sur la biomasse) ainsi qu'à son traitement ultérieur (valorisation, stockage...) (Chapitre 2, sections 3.1, 4.1).

Concernant l'évaluation du potentiel d'émissions négatives, les résultats illustrent que l'on peut obtenir un score négatif sur le changement climatique avec la substitution et l'allocation (fixation de CO_2), tandis que le score obtenu avec l'extension des frontières du système sur le système dans son ensemble est positif (émission de CO_2). Si l'ensemble du système de production ne génère pas d'émissions négatives, une augmentation du volume de production du produit auquel des émissions négatives sont attribuées n'entraînera pas une diminution du CO_2 dans l'atmosphère, au contraire. Les émissions négatives attribuées à un produit donné ne feront que compenser en partie les émissions de GES attribuées aux autres coproduits du système. En conséquence, seule l'extension des frontières du système est compatible avec l'évaluation du potentiel d'émissions négatives. Des recherches supplémentaires doivent être menées pour trouver une méthode permettant de résoudre la multifonctionnalité dans un cadre réglementaire de comptabilité carbone sans surestimer les avantages environnementaux des produits (Chapitre 2, sections 3.2 et 4.2).

Utiliser l'approche « +1/-1 » pour la comptabilisation du CO_2 atmosphérique est réalisable, mais compliqué pour les étapes d'alimentations humaine et animale du fait d'un manque actuel de données. De plus l'hypothèse d'une symétrie dans la réponse du système climatique entre les émissions et la capture du CO_2 doit être vérifiée. Si elle n'est pas valide, un facteur de caractérisation spécifique pour le captage du CO_2 atmosphérique devrait être calculé (Chapitre 2, section 4.3).

Recommandations (Chapitre 2, section 5)

Pour évaluer les systèmes CCUNET, il est essentiel de comptabiliser intégralement le carbone atmosphérique dans l'inventaire et l'évaluation de l'impact. Pour comparer les systèmes CCUNET avec d'autres technologies à émissions négatives, l'unité fonctionnelle « traitement du CO_2 atmosphérique » doit être utilisée comme fonction commune et la multifonctionnalité doit être prise en compte par l'extension des frontières du système. Le calcul du potentiel d'émissions négatives nécessite une ACV attributionnelle avec des limites de système « du berceau à la tombe » et une extension des frontières du système pour tenir compte de la multifonctionnalité.

Discussion et conclusion du chapitre

Le cas d'étude n'inclut pas de changement d'utilisation des terres. Pour un système engendrant un changement d'utilisation des terres, l'utilisation alternative des terres ne doit pas être incluse dans le système de référence, car ce n'est pas une fonction du système. L'impact du changement de l'utilisation des terres doit être pris en compte grâce à deux types de flux : les flux d'occupation et de transformation, qui seront utilisés pour calculer l'impact du système dans la catégorie d'impact « utilisation des terres » (qualité des sols, biodiversité, séquestration perdue, etc.), et les flux de polluants tels que les GES (CO_2, CH_4...), dus à la transformation directe des terres (utilisation de machines, destruction de la biomasse) (Chapitre 2, section « Land use change »).

Le cas d'étude se concentre sur l'incinération en fin de vie. Les autres options sont la mise en décharge et le recyclage. L'évaluation de l'impact environnemental de la mise en décharge est pour l'instant limitée par un manque de connaissances concernant la dégradation du plastique et l'impact des microplastiques sur la santé humaine et les écosystèmes. L'évaluation du potentiel d'émissions négatives des systèmes incluant du recyclage en boucle fermée ne soulève pas de nouveaux défis méthodologiques. Une quantité adéquate de recyclage peut même permettre de trouver un équilibre entre générer des émissions négatives et limiter les transferts d'impacts. Cependant, l'utilisation de l'extension des frontières du système soulève un défi pour l'évaluation des émissions négatives dans le cas d'un recyclage en boucle ouverte : celui de définir des limites pertinentes du système (Chapitre 2, section « End-of-life »).

Dans ce chapitre, trois recommandations spécifiques à l'évaluation des systèmes CCUNET sont formulées pour être utilisées en complément des guides méthodologiques existants pour l'ACV des systèmes CCU (1. approche « +1/-1 » pour la comptabilisation du CO_2 atmosphérique, 2. « traitement du CO_2 atmosphérique » comme unité fonctionnelle commune avec les NET, et 3. extension des frontières du système pour gérer la multifonctionnalité). Un point a été laissé pour une étude future : la dimension temporelle des émissions et des captures de CO_2 atmosphérique. Ce sujet est traité dans le chapitre suivant. L'ACV dynamique est effectuée uniquement pour la catégorie d'impact « changement climatique ». Cependant, il convient de noter que les systèmes CCUNET, même si pertinents en termes de changement climatique, peuvent également générer des transferts d'impacts significatifs.

CHAPITRE 3 : FACILITER L'EVALUATION DYNAMIQUE DU CYCLE DE VIE POUR L'ATTENUATION DU CHANGEMENT CLIMATIQUE

La question de la dimension temporelle des émissions et des captures de CO_2 est critique non seulement pour les NET mais aussi pour les systèmes CCU. L'ACV dynamique offre deux avantages majeurs (Introduction, section 4.3). Tout d'abord, elle fournit une comptabilité transparente et précise de la dimension temporelle des émissions et des captures de CO_2 atmosphérique. Deuxièmement, elle offre la possibilité de représenter l'impact sur le changement climatique sur plusieurs horizons temporels, permettant de visualiser à la fois les impacts court et long termes. L'ACV dynamique n'est pas encore disponible dans les logiciels d'ACV conventionnels et son usage n’est pas encore répandu. Cela conduit à la question traitée dans le troisième et dernier article : comment pouvons-nous faciliter l'ACV dynamique ? Trois objectifs sont abordés : les outils de modélisation (Chapitre 3, section « Modelling tool »), la dimension temporelle dans l'unité fonctionnelle et la contribution de la dimension temporelle à la précision des résultats (Chapitre 3, sections 1 et 2). Ce chapitre est en cours de soumission au journal *Sustainable Production and Consumption*: Duval-Dachary S, Lorne D, Batôt G., Helias A, Facilitating dynamic life cycle assessment for climate change mitigation.

Matériel et méthode

L'étude de cas comprend la production de biomasse (miscanthus ou résidus de bois), la fermentation de la biomasse, la capture du CO_2 et sa valorisation en sac en polypropylène, et l'incinération du sac avec capture et stockage du CO_2. Le miscanthus est une culture pérenne (non replantée chaque année) et donc capable de générer un stockage de carbone dans le sol grâce à la croissance de son réseau racinaire. Les résidus de bois ne sont pas les produits principaux du système et sont en général laissés sur place. Les résidus se voient attribuer des émissions et une consommation de ressources uniquement pour les étapes nécessaires à leur transformation en produits valorisables (par exemple, collecte, séchage, etc.).

L'ACV dynamique consiste à intégrer des informations temporelles dans l'inventaire du cycle de vie, c'est-à-dire indiquer pour chaque flux vers ou de l'environnement l'instant d'émission ou de capture. On a donc une chronologie, ou distribution temporelle, d'émissions et de captures constituant l'inventaire dynamique du cycle de vie. Le pas de temps à utiliser dépend de la catégorie d'impact. Pour le changement climatique, un pas de temps d'un an minimum est suffisant. Les facteurs de caractérisation utilisés pour convertir les masses émises ou capturées en impact potentiel sur l'environnement dépendent alors non seulement de la substance émise ou capturée, mais également de son instant d'émission ou de capture. Il faut également définir un horizon temporel pour lequel l'impact potentiel est calculé (classiquement 20, 100 ou 500 ans pour le changement climatique). Cela signifie définir un instant zéro (t_0) à partir duquel l'horizon temporel est comptabilisé et autour duquel il faut positionner la distribution temporelle de l'inventaire dynamique.

Les distributions temporelles de production des biomasses (production sur 15 ans pour le miscanthus ou 180 ans pour les résidus de bois) ne sont pas égales à leurs distributions temporelles de consommation dans le processus de production d'éthanol (quantité fixe tous les ans pendant 20 ans). Un algorithme est donc proposé pour moyenner temporellement les inventaires de production de biomasse (Chapitre 3, section 3.1). Temporalis, une librairie de Brightway2 en Python, a été modifiée et utilisée pour calculer l'inventaire dynamique et la caractérisation dynamique de l'impact (Chapitre 3, section 3.2).

Deux unités fonctionnelles sont comparées : « quantité totale d'unités produites répartie sur toute la durée de vie de l'installation » (UF_1) et « quantité totale produite à t_0 » (UF_2). Deux positionnements temporels de l'inventaire par rapport à l'instant zéro de la caractérisation de l'impact sont également comparés pour UF_1 : première année de production égale à l'instant zéro ($P_{début} = t_0$) ou dernière année de production ($P_{fin} = t_0$)(Chapitre 3, section 3.3).

Une analyse de sensibilité (indices de Sobol) est réalisée pour identifier les paramètres contribuant le plus à la variabilité des résultats en statique. Les variations engendrées par ces paramètres sont ensuite mises en regard des variations engendrées par la modélisation en dynamique, en utilisant l'UF_2 pour limiter le nombre de paramètres temporels (Chapitre 3, section 3.4).

Résultats

Temporalis, l'outil actuellement disponible pour réaliser des ACVs dynamiques, a été testé et amélioré. Tous les documents seront mis à disposition avec l'article pour faciliter les futures réalisations d'ACVs dynamiques (Chapitre 3, section 4.1).

Les résultats obtenus avec l'UF$_1$ sont dans un intervalle de ± 5% des résultats obtenus avec l'UF$_2$ pour des horizons temporels très supérieurs à la durée de vie de l'usine. Le cas d'étude est représentatif d'un cas particulier où le même inventaire dynamique est utilisé pour modéliser la production d'une unité pour l'UF$_1$ et l'UF$_2$. Cela correspond au cas où il n'y a pas d'important pic d'émissions dû à la production et déconstruction des infrastructures par exemple. Dans ce cas, quand l'horizon temporel tend vers l'infini, l'impact calculé avec UF$_2$ est la moyenne de l'impact calculé avec UF$_1$ ($P_{début} = t_0$) et UF$_1$($P_{fin} = t_0$)(Chapitre 3, section 4.2).

Les deux paramètres qui sont responsables de la majorité de la variation des résultats en statique sont le stock initial de carbone dans le sol et la modélisation de la production d'énergie (chaleur et hydrogène)(Chapitre 3, section 4.3.1). L'impact de la modélisation dynamique de la modification du stock de carbone dans le sol lors de la croissance du miscanthus est négligeable par rapport aux variations induites par la méconnaissance du stock initial de carbone dans le sol. Par contre, la modélisation dynamique des résidus de bois engendre une variation par rapport au statique du même ordre de grandeur que la variation induite par la modélisation de la production d'énergie (pour un horizon de 100 ans). Quand l'horizon temporel tend vers l'infini, l'écart entre l'impact du CO$_2$ calculé en dynamique et celui en statique tend vers $a_{CO2}a_0 \sum_{te} m_e t_e$, avec a_{CO2} l'efficacité radiative instantanée du CO$_2$, a_0 le premier coefficient de la fonction de dégradation du CO$_2$, et m_e la masse de CO$_2$ émise au temps t_e (Chapitre 3, section 4.3.2).

Discussion et conclusion

Temporalis est un outil efficace pour réaliser une ACV dynamique. Deux pistes d'amélioration ont été identifiés : le traitement de la perte d'informations due à la condition d'arrêt de l'algorithme utilisé pour calculer l'inventaire et l'augmentation du nombre de méthodes de caractérisation proposées pour pouvoir effectuer une analyse de sensibilité. La version opérationnelle de Temporalis, l'algorithme pour moyenner un inventaire dynamique, et l'exemple réutilisable fourni dans ce chapitre faciliteront l'utilisation de l'ACV dynamique (Chapitre 3, section 5.1).

L'UF$_1$ doit être utilisée pour évaluer l'impact potentiel sur le changement climatique de l'ensemble du système et le mettre en relation avec des objectifs climatiques à atteindre à une date précise (ex : neutralité carbone à 2050). Pour comparer les résultats obtenus aux résultats statiques, l'horizon temporel doit être défini à partir de la dernière année de production. L'UF$_2$ doit être utilisée pour comparer les systèmes qui ne partagent pas la même distribution temporelle de production et pour construire des inventaires moyens réutilisables dans d'autres cycles de vie. L'UF$_2$ peut ainsi être utilisée pour comparer de manière pertinente les systèmes CCUNET et les autres systèmes NET qui ne partagent pas les mêmes distributions temporelles pour la production de la fonction 'traitement de

CO_2 atmosphérique' (par exemple un système de reforestation) (Chapitre 3, sections 5.2 et « Chapter conclusion »).

Cependant, les résultats montrent également que l'effort nécessaire pour réaliser une ACV dynamique ne conduit pas nécessairement à des résultats très différents de ceux de l'ACV statique (exemple du miscanthus). Les propriétés mathématiques du potentiel de réchauffement global absolu (GWP en anglais) pour un horizon temporel tendant vers l'infini ont été étudiées, permettant de prédire certains des résultats en utilisant uniquement des informations temporelles simplifiées. Une méthode est nécessaire pour permettre aux praticiens de l'ACV de cibler plus efficacement leurs efforts, c'est-à-dire leur permettre d'identifier à l'aide d'informations temporelles simplifiées les flux pour lesquels l'ajout d'informations temporelles est crucial. C'est le sujet du chapitre suivant.

CHAPITRE 4 : L'ACV DYNAMIQUE EST-ELLE NECESSAIRE ? EVALUATION AVEC DES INFORMATIONS TEMPORELLES SIMPLIFIEES

Un praticien de l'ACV dispose d'un temps limité et peut être amené à choisir entre l'amélioration de ses données d'inventaire dans le cadre d'une ACV statique et la réalisation d'une ACV dynamique. Le chapitre précédent montre que l'ACV dynamique exige davantage de données sur le système et accroît la complexité des calculs, mais que la précision des résultats n'est pas nécessairement améliorée de manière significative. D'où la question soulevée dans ce chapitre : en se concentrant sur le changement climatique, est-il possible de déterminer si l'approche dynamique modifiera significativement les résultats par rapport à une approche statique avec seulement une connaissance limitée de la dynamique du système étudié ? Pour répondre à cette question, le problème est d'abord exprimé mathématiquement. Ensuite, les expressions mathématiques sont calculées pour le potentiel de réchauffement global absolu. Puis, la méthode est appliquée à quelques exemples. Et enfin des valeurs seuils de l'amplitude temporelle à partir de laquelle l'ACV dynamique est nécessaire sont fournies pour un nombre limité de profils d'émissions.

Expression mathématique du problème

L'objectif d'une ACV n'est pas seulement de calculer un score final, mais aussi de connaître les principaux contributeurs. Substance par substance, les résultats calculés à l'aide d'une approche dynamique (I_{dyn}) peuvent être comparés aux résultats obtenus à l'aide d'une approche statique (I_{stat}) en examinant le rapport entre ces résultats, c'est-à-dire I_{dyn}/I_{stat}. Plus ce rapport est proche de un, moins l'approche dynamique est intéressante (Chapitre 4, section 1) .

$$\left[\frac{I_{dyn}}{I_{stat}}\right]_i = \sum_{t_e}\left[\frac{m_i(t_e)}{m_i^{total}} \times \frac{CF_i(TH - t_e)}{CF_i(TH)}\right]$$

Avec:

- $\frac{m_i(t_e)}{m_i^{total}}$ le rapport de masse entre la quantité de GES$_i$ émise à l'instant t_e et sa quantité totale émise sur l'ensemble du cycle de vie. Il représente la distribution des émissions dans le temps. Des motifs d'émissions simples, « pic » et « linéaire », sont proposés pour permettre le calcul de ce rapport en utilisant le moins d'informations temporelles possible. Si le profil d'émissions est symétrique autour de t_0, et que le rapport des facteurs de caractérisation est une fonction linéaire de l'instant d'émission, le rapport I_{dyn}/I_{stat} est égal à 1 (Chapitre 4, section 1.1).
- $\frac{CF_i(TH-t_e)}{CF_i(TH)}$ le rapport du facteur de caractérisation dynamique d'une émission à t_e sur le facteur de caractérisation statique pour le même horizon temporel TH. Il correspond à la variation du facteur de caractérisation due à la prise en compte de la temporalité. Ce ratio est calculé numériquement, à l'aide d'un script python. Pour le potentiel de réchauffement global absolu du CO_2 et du CH_4 ce ratio est proche d'une fonction linéaire de t_e si celui-ci est compris entre $-40\%TH$ et $40\%TH$ (Chapitre 4, section 2).

Résultats

Cette méthode permet de retrouver facilement les conclusions du chapitre précédent. La variation moyenne de stock de carbone dans le sol due à la plantation de miscanthus peut s'approximer par un profil d'émissions symétrique autour de t_0. Le ratio I_{dyn}/I_{stat} est donc égal à 1, il n'est pas nécessaire d'utiliser l'ACV dynamique pour modéliser la variation de stock de carbone dans le sol. Pour les résidus de bois, la capture de CO_2 peut s'approximer par une fonction linéaire décroissante entre -180 ans et t_0. Le ratio I_{dyn}/I_{stat} n'est pas compris entre 0.9 et 1.1, l'ACV dynamique est nécessaire (10% est l'incertitude conventionnelle associée aux facteurs de caractérisation pour le changement climatique). Cette méthode est appliquée à d'autres exemples provenant de la littérature : production d'énergie par panneau solaire et émissions d'une décharge (Chapitre 4, section 3).

Un type de profil d'émission semble très commun : la fonction linéaire croissante ou décroissante dont l'une des dates extrêmes est t_0, et l'une des masses extrêmes vaut zéro. Le ratio I_{dyn}/I_{stat} ne dépend alors que de deux informations temporelles : la seconde date extrême, notée $t_{extrême}$, ainsi que le type de profil (« linéaire avec $m(t_{extrême}) = 0\,kg$ » ou « linéaire avec $m(t_0) = 0\,kg$ »). C'est également le cas pour les profils « pic » et les profils « uniforme » dont l'une des dates extrêmes est t_0. Pour ces quatre types de profils, il est possible de proposer des valeurs seuils en dehors desquelles l'ACV dynamique est nécessaire. Par exemple, quel que soit l'horizon temporel, si $t_{extrême}$ d'un profil « linéaire avec $m(t_{extrême}) = 0\,kg$ » est de l'ordre de ± 40% du TH, le ratio I_{dyn}/I_{stat} est compris entre 0.9 et 1.1 : l'ACV dynamique n'est pas nécessaire (Chapitre 4, section 4).

Conclusion

Dans ce chapitre, des valeurs seuils de durée de motif sont fournies, permettant aux praticiens de l'ACV d'identifier si l'ACV dynamique est nécessaire uniquement avec deux informations temporelles : le type de profil d'émission (« pic », « linéaire avec $m(t_{extrême}) = 0\,kg$ », « linéaire avec $m(t_0) = 0\,kg$ », « uniforme ») et la durée du motif ($t_{extrême}$). Si le profil d'émission est plus complexe ou si le praticien de l'ACV dispose de plus d'informations sur l'incertitude des résultats statiques et souhaite utiliser un seuil plus bas ou plus élevé pour décider si l'ACV dynamique est nécessaire, le ratio I_{dyn}/I_{stat} peut être calculé numériquement. Ces valeurs seuils d'amplitude temporelle pourraient être calculées pour plus de GES et pour d'autres indicateurs du changement climatique dans des travaux futurs.

CONCLUSION GENERALE ET PERSPECTIVE

Au cours des quatre chapitres, des contributions méthodologiques et pratiques ont été apportées pour faciliter et améliorer l'évaluation des émissions négatives des systèmes CCU. Les contributions méthodologiques comprennent les recommandations proposées dans le chapitre 2 (comptabilisation du CO_2 atmosphérique, définition de l'unité fonctionnelle, méthode pour gérer la multifonctionnalité), dans le chapitre 3 (unité fonctionnelle dynamique), ainsi que la méthode proposée dans le chapitre 4 pour permettre aux praticiens de l'ACV de cibler plus efficacement leurs efforts en identifiant à l'aide d'informations temporelles simplifiées les flux pour lesquels l'ajout d'informations temporelles est crucial (Conclusion générale, section 1.1). Les contributions pratiques comprennent les tables d'inventaires compilées dans le chapitre 1 pour faciliter l'utilisation des frontières « du berceau à la tombe », le facteur de conversion de la masse de carbone ingérée en masse émise de CO_2 et CH_4 pour une étape d'alimentation de ruminants proposés dans le chapitre 2, et tous les scripts python fournis pour permettre la reproductibilité de l'étude, mais également fournir des exemples d'utilisation des outils émergents (Brightway2, Temporalis) (Conclusion générale, section 1.2).

Des lacunes dans la littérature scientifique ont été identifiées tout au long de ce travail, en ce qui concerne les données d'inventaire, la caractérisation de l'impact et l'ACV en tant qu'outil de prise de décision dans le contexte des systèmes CCU et/ou NET. Les perspectives de recherche sont donc les suivantes :

- Concernant les données d'inventaires, d'un point de vue opérationnel, il serait intéressant de créer une base de données open-source pour un partage de données plus efficace au sein des utilisateurs de Brightway2 (Conclusion générale, section 2.1).
- Ensuite, la caractérisation de l'impact du changement climatique peut être améliorée en vérifiant l'hypothèse d'une symétrie de réponse du système climatique par rapport à la capture et

l’émission de CO_2, et également en intégrant les effets de rétroaction CO_2-climat dans l'ACV dynamique. De manière plus générale, il serait intéressant d’inclure l'incertitude des méthodes de caractérisation dans Temporalis tout en conservant la clarté des résultats (Conclusion générale, section 2.2).

- Enfin pour faciliter la prise de décision à l’aide de l’ACV dans le contexte des systèmes CCUNET, il serait intéressant de traduire les recommandations proposées dans les chapitres 2 et 3 dans le cadre spécifique de la comptabilité carbone (affichage environnemental, marché européen du carbone) et de construire un outil d'ACV territoriale dynamique pour faciliter l'appropriation des technologies CCU/NET par les décideurs publics (Conclusion générale, section 2.3).

Scientific production

- Chapter 1 was published and presented in oral communications:
    - Duval-Dachary S., Beauchet S., Lorne D., Salou T., Helias A., Pastor A. (2023) Life cycle assessment of bioenergy with carbon capture and storage systems: Critical review of life cycle inventories. *Renewable and Sustainable Energy Reviews* 183:113415. https://doi.org/10.1016/j.rser.2023.113415
    - Duval-Dachary S., Pastor A., Beauchet S., Lorne D., Salou T. and Helias A., Life Cycle Assessment of BECCS systems: critical review of life cycle inventories (2022) https://dx.doi.org/10.2139/ssrn.4271614 *16th International Conference on Greenhouse Gas Control Technologies GHGT-16* (Lyon, France)
    - Duval-Dachary S., Pastor A., Beauchet S., Lorne D., Salou T. and Helias A., LCA of biogenic carbon capture & storage processes : review of life cycle inventories and recommendations (2023) *SETAC Europe 33rd annual meeting (Dublin, Ireland)*
- Chapter 2 was:
    - submitted to the *International Journal of Life Cycle Assessment* (and is currently being revised as): Duval-Dachary S., Lorne D., Beauchet S., Salou T., Hélias A. Life cycle assessment of carbon capture and utilisation as a negative emission technology: recommendations and case study
    - presented in oral communication: Duval-Dachary S., Pastor A., Beauchet S., Lorne D., Salou T. and Helias A., A guideline for life cycle assessment of carbon capture and utilization as negative emissions technologies (2023) *SETAC Europe 33rd annual meeting (Dublin, Ireland)*
- Chapter 3 is ready to be published as: Duval-Dachary S., Lorne D., Batôt G., Helias A, Facilitating dynamic life cycle assessment for climate change mitigation In *Sustainable Production and Consumption*
- Chapter 4 was presented in oral communication: Duval-Dachary S., Beauchet S., Lorne D., Salou T and Helias A., Result variations due to dynamic life cycle assessment compared to result variations due to sensitivity analysis on static inventory data (2024) *SETAC Europe 34th annual meeting* (Seville, Spain)

Tables

Figures

Keys definitions, acronyms and abbreviations

The definition of the main concepts mentioned in this work are summarised in this section, along with the acronyms and abbreviations used. The definitions are mostly recalled when first mentioned in the text. This section is to be used during reading to easily retrieve a definition if the reader has forgotten the meaning of a concept or acronym.

"+1/-1" approach	Full accounting of atmospheric carbon in the inventory and in the impact assessment. Emissions of atmospheric carbon (CO_2, CH_4..) are included as positive masses in the inventory, while captures are included as negative masses. The characterisation factor is the same for both capture and emission and corresponds to the characterisation factor used for fossil carbon.
(A)GWP	The (Absolute) Global Warming Potential converts emissions of GHG into impact on radiative forcing. Its formula is given in section 4.1 of the general introduction.
(A)GTP	The (Absolute) Global Temperature Change evaluates the increase in global temperature compared to the pre-industrial temperature due to GHG emissions.
Allocation	Performing allocation is a solution to deal with multifunctionality. The impact is distributed between products based on physical factors (mass, energy or carbon content) or economic factors. For instance, a system produces two products, P_1 and P_2. P_1 has a carbon content of c_1 (in kgC/kg) and P_2 of c_2. The impact of producing m_1 kg of P_1 and m_2 kg of P_2 is I_{total}. The impact allocated by carbon content to the production of the m_1 kg of P_1 will be $\frac{m_1 c_1}{m_1 c_1 + m_2 c_2} I_{total}$.
Attributional LCA	The objective of an attributional LCA is to evaluate the environmental impact that can be associated with a product or service production. It is assumed that the background system is not modified by the studied system. Typically, in attributional LCA the average electricity mix is used for modelling electricity consumption.

BECCS	Bioenergy with carbon capture and storage. BECCS are systems that convert biomass into energy and capture the CO_2 produced for permanent storage in geological reservoirs.
Brightway2	“Open-source software package for life cycle assessment (LCA) and environmental impact assessment written in the Python programming language” (https://docs.brightway.dev/en/latest/)
Carbon removal	See Negative emissions.
CF	Characterisation factors. CF links the emissions/consumptions listed in the LCI to their potential impact on the environment (e.g. Global Warming Potential).
Circular footprint formula	The CFF was developed to allocate the burdens and credits of recycling between supplier of waste and user of recycled raw materials by the European Union. It is described in Annexe 1.
Consequential LCA	The objective of a consequential LCA is to evaluate the environmental impact of changes due to decisions or variations in demand/supply. The flows modelled in the LCI included all the flows that vary between the baseline scenario and the evaluated scenario. Typically, in consequential LCA the electricity consumption is modelled by the mode of production affected by the increase in electricity consumption (marginal mode of electricity production).
CCU	Carbon Capture and Utilisation. CCU consists of recycling captured CO_2, with or without transformation, for further usage, such as fuel or plastic production. According to the European Union, CCU can contribute to energy security, emission reduction, and autonomy.
CCUNET	Carbon capture and utilisation as a negative emissions technology.
DACCS	Direct air capture with carbon capture and storage. DACCS refers to technologies that remove dilute CO_2 from the surrounding atmosphere for permanent storage in geological reservoirs.
Dynamic LCA	LCA studies where flows from and to the environment are distributed on a time scale (temporal differentiation) to then apply characterisation factors that depends on the time of capture or emission. For a full description see section 4.3 of the general introduction.

Emissions pattern	Succession of pulse emissions that can be described by a mathematical function.
FU	The functional unit is the common point of comparison of the systems. For example, to compare two means of passenger transport, it can be "the transport of one person over 1 km".
GHG	Greenhouse gases (water vapour, CO_2, CH_4…) induce warming by increasing the greenhouse effect.
LCA	Life cycle assessment. The standardised method for evaluating the environmental impact of produce and service. See section 4 of the general introduction for full description.
LCI	Life cycle inventory. The LCI assembles all emissions to the environment as well as the resource consumption from the environment caused by the system under study.
LCIA	Life cycle impact assessment. Conversion of the LCI into potential impacts on the environment thanks to characterisation factors.
Multifunctionality	A system can have multiple products (ex: a refinery) and/or multiple functions (ex: passenger and freight transport). To make a comparison it is sometimes necessary to isolate the impact of one of the functions or products.
Negative emissions	Negative emissions are defined by Minx et al. (2018) as “intentional human effort to remove CO_2 emissions from the atmosphere”. Tanzer and Ramirez (2019) specified the requirements to achieve net negative emissions: i) greenhouse gases (GHG) must be removed from the atmosphere and stored; ii) the GHG emissions over the whole life cycle of the system must not offset the amount of GHG removed.
NET	Negative Emissions Technology.
Substitution	Substitution is a solution to deal with multifunctionality. The impact of one of the system functions is isolated by subtracting the impact of the production of the unwanted products calculated using single-output processes. Substitution potentially leads to negative results.

System boundaries	They encompass all steps of the life cycle that will be considered in the assessment. “From-cradle-to-grave” system boundaries, refers to systems boundaries including all steps, from material extraction to the end-of-life of the product, from CO_2 capture from the atmosphere to its permanent storage out of the atmosphere or its release.
System expansion	System expansion is a solution to deal with multifunctionality. The system boundaries are extended until they include the complete life cycle of each co-product and if needed additional single-output processes to match the functions of the compared system.
Temporalis	Python-based library to perform dynamic LCA using Brightway2.
Temporal differentiation	“The action of distributing the information on a time scale related to the models' components. For example, elementary flows could be described per day or year.” (Beloin-Saint-Pierre et al. 2020)
Temporal scope	“Defines any type of period that is considered in a LCA study (e.g. temporal considerations along a life cycle, service life of a product, data collection period).” (Beloin-Saint-Pierre et al. 2020)
TH	Time horizon. Duration of the impact assessment. Usually 20, 100 or 500 years for the GWP.

General Introduction

The global crisis of climate change and the pursuit of carbon neutrality, presented in section 1, revealed a need for removal of carbon dioxide from the atmosphere, using negative emissions technologies (NET), presented in section 2. Compared to carbon removal, carbon capture and utilisation (CCU) transform the CO_2 into valuable products, replacing fossil raw materials and generating business opportunities, so coupling CCU and NET is beginning to attract interest, as introduced in section 3. Evaluating the environmental relevance of CCU as NET systems requires an assessment over their entire lifecycle, using life cycle assessment (LCA). LCA methodology is quite mature. However, some challenges remain, such as the temporal limitations of LCA. This topic is covered in section 4. Section 5 concludes this introduction with the presentation of this thesis research question and the associated research strategy to answer it.

1 CLIMATE CHANGE: HOW? WHY? WHAT? WHEN?

According to the IPCC (2021), the role of the atmosphere in maintaining a liveable temperature on earth, through the greenhouse effect, is established since the 19th century. The atmosphere's capacity to absorb the radiation emitted by the Earth’s surface depends on its concentration of radiative forcers. Variation of the concentration of radiative forcers causes changes of the Earth’s energy budget. The Earth’s energy budget is the difference between the energy absorbed from solar radiation and the energy re-radiated as heat to space. Radiative forcers include greenhouse gases (GHG) (water vapour, CO_2, CH_4…), inducing warming by increasing the greenhouse effect, and aerosols, inducing cooling by increasing the amount of solar energy reflected into space. The net change in the energy budget is called radiative forcing (in W/m^2). An increase of radiative forcing causes an increase of the Earth average surface temperature: the global warming.

Human activities cause anthropogenic emissions of GHG. For instance, the atmospheric CO_2 concentration increased by almost 50% over the period 1750-2019 due to emissions of CO_2 from the burning of fossil fuels and industrial processes. Thus, according to the latest IPCC report (IPCC 2023), “Human activities […] have unequivocally caused global warming, with global surface temperature reaching 1.1°C above 1850-1900 in 2011-2020”.

Global warming has short- and long-term consequences. Short-term consequences include increasing frequency of extreme weather events, such as floods, drought, and storms. It has an impact on water and food security but also on human health (disease, mental health, trauma) and biodiversity (IPCC 2023). Long-term consequences include permafrost melting or sea-level rises, due to the retreat of glaciers and to ocean warming (water expansion)(IPCC 2023). Already “3.3 to 3.6 billion people live in contexts that are highly vulnerable to climate change” (IPCC 2023). In response to this global crisis, 196 nations signed the Paris agreements in 2015. They decided to limit global warming to 2°C, or even 1.5°C (UNFCCC). Stabilizing human-induced global warming requires reaching net zero CO_2 emissions, and a decrease in non-CO_2 forcing emitted by human activities (IPCC 2021), before overshooting the remaining carbon budget. According to the IPCC (2021), the remaining carbon budget to limit global warming to 1.5°C is 500 $GtCO_2$ starting from 1 January 2020. In 2019, “global net anthropogenic GHG emissions have been estimated to be 59 ± 6.6 $GtCO_2eq$”. Ambitious strategies are therefore needed in all countries to drastically and rapidly reduce anthropogenic GHG emissions. In the following section, the mitigation strategies of the European Union and France are presented.

2 MITIGATION STRATEGIES OF CLIMATE CHANGE OF EUROPE AND FRANCE: WHICH ROLE FOR NEGATIVE EMISSIONS AND CARBON CAPTURE AND STORAGE?

The ambition of the European Union is to reduce GHG emissions by at least 55% by 2030 compared to their 1990 levels and to achieve carbon neutrality in 2050 (European Council 2023). The European Union's main tool in addressing emissions reductions is the European Union's emissions trading system, a carbon market with annual emissions cap. This ensures that polluters pay. Moreover, the European Union plans a transition toward renewable energy and decarbonized fuels, accompanied by an increase in energy efficiency. They also assert the need for atmospheric carbon removal before 2040 to first counterbalance hard-to-abate emissions and then achieve negative emissions, in their strategy "towards an ambitious Industrial Carbon Management" (European Commission 2024). They aim to reach a net removal of 310 million tonnes CO2eq in 2030 through the land use, land use change and forestry sector. They recognise the potential need for technical solutions such as bioenergy with carbon capture and storage (BECCS), but have not set a quantitative target for deployment (European Parliament and Council 2023). In France, an additional lever is used: sufficiency (e.g. managing demand growth, changing lifestyles and consumption...) (MTES 2020). France's ambition is also to reach carbon neutrality in 2050. However, using only the lever "avoid" and "reduce", around 80 MtCO2eq are still emitted per year in 2050 by the agricultural sector and industrial processes. Therefore, France recognises the need for atmospheric carbon removal in order to offset hard-to-abate residual GHG emissions and reach carbon neutrality in 2050.

Negative emissions, or carbon removal, is defined by Minx et al. (2018) as "intentional human effort to remove CO_2 emissions from the atmosphere". The current focus of the scientific community is on technologies removing CO_2 from the atmosphere, but Minx et al. (2018) "note the existence of technologies that remove other non-CO_2 greenhouse gases from the atmosphere". Minx et al. (2018) proposed a taxonomy of negative emissions technologies (NET). They identified three main parameters: the type of capture (photosynthesis or chemistry), the earth system involved (land or ocean) and the long-term storage medium (biomass, soil, geological reservoirs, minerals, marine sediment & calcifiers). There are about ten different technology clusters of NET, but the European Union and France focuses on carbon storage in soils and forests, BECCS, and direct air capture and carbon storage (DACCS) (European Council 2024; MTES 2020). Carbon storage in soils and forests is a low-cost solution but with a limited capacity (stock saturation) and a risk of storage reversibility (e.g. forest fire) (Fuss et al. 2018). BECCS are systems that convert biomass into energy and capture the CO_2 produced for permanent storage in geological reservoirs. BECCS faces competition for the use of biomass and land, also needed by the food and feed sector. DACCS refers to technologies that remove dilute CO_2 from the surrounding atmosphere for permanent storage in geological reservoirs. The current cost is high, around \$600/tCO_2 (Fuss et al. 2018). Geological storage is a reliable storage option with low risk of reversibility (Fuss et al. 2018).

The European Union also include carbon capture and utilisation (CCU) in its industrial carbon management (European Commission 2024). CCU consists of recycling captured CO_2, with or without transformation, for further usage, such as fuel or plastic production. According to the European Union, CCU can contribute to energy security, emission reduction, and autonomy. The European chemical industry uses approximately 125 Mt of carbon, 90% of which comes from fossil resources (European Commission 2024). France also include CCU in its strategy, predominantly for producing fuels (aviation and maritime) and also for long-term storage in products (concrete carbonation). Compared to carbon capture and storage, CCU provides economic opportunities and its deployment is less hampered by the need for CO_2 transportation (MTES 2020). The European Union is preparing to recognise the permanent storage of carbon in products under certain conditions (European Commission 2024). In the following section, the CCU systems with potential for negative emissions are explored.

3 IS IT POSSIBLE TO GENERATE NEGATIVE EMISSIONS WITH CCU?

There are various uses of the CO_2, with or without (chemical or biological) transformation (Kerlero de Rosbo et al. 2014). The relevance of CCU systems for short-term deployment has been discussed in the literature, as shown in Table 1. Methanol is a major building block in the chemical industry along with ethylene, propylene, BTX (benzene, toluene, xylene), ammonia, urea and chlorine (Bazzanella and Ausfelder 2017). As shown in Table 1, there is a consensus on the potential of producing methanol from CO_2. Methanol can either be used directly as a fuel or converted into a wide range of products usually derived from petrochemicals. For urea and concrete curing, visible in the Table 1, the CO_2 is more likely to be fossil CO_2 produced internally by the industry (ammonia production and limestone decarbonation) (Kerlero de Rosbo et al. 2014, Cembureau 2020).

Table 1: The most promising CCU systems identified in (CarbonNext 2018; Chauvy et al. 2019; Kerlero de Rosbo et al. 2014; Otto et al. 2015; Patricio et al. 2017) are indicated by a cross (x).

CCU systems	x: most promising CCU system identified in the articles				
	(Otto et al. 2015)	(Chauvy et al. 2019)	(Kerlero de Rosbo et al. 2014)	(CarbonNext 2018)	(Patricio et al. 2017)
Methanol	x	x	x	x	x
Ethylene and propylene				x	x
Benzene, Toluene, Xylenes				x	
Ammonia and urea	x				x
Dimethyl ether	x			x	
Organic Carbonate (ethylene, dimethyl…)		x		x	x
Formic acid	x		x		
Oxalic acid	x				
Formaldehyde	x				
Lignin production					x
Polyurethane (polyol, carbamate…)					x
Concrete curing					x
Bauxite residue carbonation					x
Mineral carbonation		x	x		x
Energy (methane, fuels…)		x		x	
Acrylate					
Carbon monoxide					

Desport and Selosse (2022) present a comprehensive review of CCU systems capable of achieving net negative emissions and refer to them with the acronym CCUNET: carbon capture and utilisation as a negative emissions technology. In Figure 1, the general life cycle of a CCUNET system is illustrated. It can be divided in three main steps: producing the CO_2, valorising the CO_2 and using the CO_2-based product, and lastly treating the CO_2-based product at its end-of-life. To obtain negative emissions, the valorised CO_2 must initially come from the atmosphere. Thus, in Figure 1, two options are represented for the system producing the CO_2: the CO_2 is either captured in the flue gas of an industry that uses biomass as a raw material, or directly from the atmosphere by direct air capture. Negative emissions can only be generated when CO_2 is kept out of the atmosphere for at least several decades. The CO_2 uses that are compatible with negative emissions are therefore those that allow permanent storage in long life products (concrete, mineralisation, etc.), by "infinite" reuse (solvent, supercritical cycle, reuse and recycling of the product, etc.), or at the end of the product's life (landfill, storage in geological reservoir, etc.). Thus, in Figure 1, the three end-of-life that may lead to negative emissions are represented: landfill, incineration with carbon capture and storage and recycling.

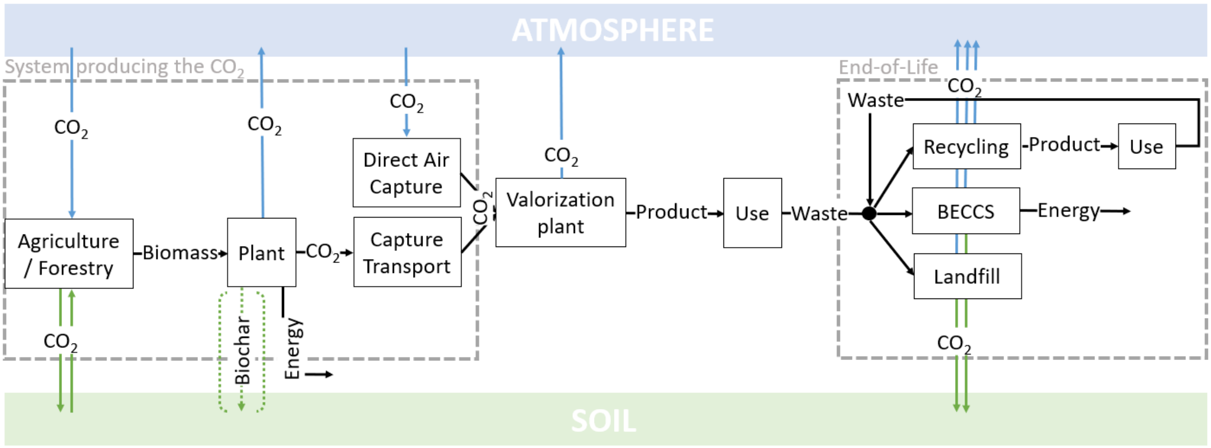

Figure 1: Life cycle of atmospheric carbon capture and utilisation system with potential to generate negative emissions. Only exchanges of CO_2 between the system and the environment are shown to make the figure readable, but there are obviously other resources consumed and pollutants emitted. BECCS: Bioenergy with carbon capture and storage.

Tanzer and Ramirez (2019) specified the requirements to achieve net negative emissions: i) GHG must be removed from the atmosphere and stored; ii) the GHG emissions over the whole life cycle of the system must not offset the amount of GHG removed. CCUNET systems as defined in Figure 1 fulfil the first condition: CO_2 is removed from the atmosphere and stored. However, it is more complex to check if CCUNET system fulfil the second condition, as it requires an assessment over the entire life cycle. Indeed, the production of energy and chemicals that are consumed during processes (e.g. CO_2 capture) leads to additional emissions of GHG. CCUNET systems may also be responsible for other types of environmental impacts, such as acidification, water footprint or human toxicity (Oreggioni et al. 2017). Moreover, CCU systems generate avoided emissions that are also complex to quantify. Products created from CCU processes are substituted for products generally created from raw materials of fossil origin (reference products). The environmental benefits of this substitution, i.e. the avoided emissions, are then assessed by comparing the impact of the production of the reference product (e.g. gasoline) with the impact of the production of the CCU-based product (e.g. synthetic fuel). It can be difficult to assess avoided emissions, as the reference product differs by location and may change over time (e.g. as the energy mix of the transport sector becomes less dependent on fossil fuels). Quantifying these benefits (both avoided and negative emissions) is therefore complex and requires assessment over the entire life cycle of the product. The life cycle assessment (LCA) methodology and its challenges are covered in the next subsection.

4 EVALUATING NEGATIVE EMISSIONS POTENTIAL REQUIRES LIFE CYCLE ASSESSMENT (LCA)

LCA is the standardised method to perform the environmental assessment of products and services. According to the ISO standard 14040-14044 (2006a; 2006b), LCA is performed in four steps:

1. Goal and scope definition. In this step, the questions, which will be answered by the LCA, are defined. For instance, it can be: ‘Is product A environmentally better than product B?’. Then the methodological choices and main assumptions are explicitly given to define the scope within which the results of the LCA are valid. It notably includes:
    a. the boundaries of the system under study. This includes each step of the life cycle that will be considered in the assessment, from material extraction to the end-of-life of the product, but also geographical and temporal boundaries (for instance, the product A produced in France between 2020-2022).
    b. the functional unit (FU) that will be the common point of comparison of the systems. For example, to compare two means of passenger transport, it can be "the transport of one person over 1 km".
    c. the method to solve multifunctionality if needed. A system can have multiple products (ex: a refinery) and/or multiple functions (ex: passenger and freight transport). To make a comparison it is sometimes necessary to isolate the impact of one of the functions or products. The results can be highly dependent of the method chosen to solve multifunctionality.
    d. information about life cycle inventory data quality.
2. Life cycle inventory (LCI). In this step, data is collected to calculate the LCI of the system. An LCI assembles all emissions to the environment as well as the resource consumption from the environment caused by the system under study. Collecting data to build the LCI is an essential but time-consuming part of conducting an LCA. Data comes from various sources (system modelling, field measurement, literature, database).
3. Life cycle impact assessment (LCIA). During this step, the LCI is converted into potential impacts on the environment thanks to characterisation factors. Multiple impact categories are considered (climate change, acidification, eutrophication…) to be able to identify potential trade-offs between categories. In static LCA, the impact I of a product or service for a specific impact category is calculated according to the following formula:

$$I(TH) = \sum_s g_s CF_s(TH) \tag{1}$$

with:

- g_s: the total amount of substances (e.g. CO_2, CH_4 ...) emitted/consumed during the life cycle,
- CF_s : the characterisation factor associated with the substance that links its emission/consumption and its impact (e.g. Global Warming Potential),
- TH: the time horizon chosen for the impact assessment (e.g. 100 years).

4. Interpretation. In this step, the contribution to the impact of substances and processes is analysed to identify errors and to judge the relevance of the modelling assumptions. For example, if a process contributes significantly to the impact, the LCA practitioner may decide to refine the LCI for that particular process. Sensitivity and uncertainty analysis are also performed. This interpretation step may lead to modification of the previous three steps and iterations until the questions raised in the goal can be satisfactorily answered.

The evaluation of negative emission potential is linked to the interpretation of LCA results in the impact category “climate change”. The methods for characterising the impact on climate change are therefore described in the subsection 4.1. As explained in section 3, to generate negative emissions, CO_2 must initially be captured from the atmosphere. The methodological challenges raised by atmospheric CO_2 accounting are explained in the subsection 4.2. The subsection 4.3 introduces the concept of dynamic LCA, one of the solutions to account for atmospheric CO_2.

4.1 CHARACTERISATION METHODS TO EVALUATE THE IMPACT ON CLIMATE CHANGE

As explained by the UNEP-SETAC (2016), radiative forcers can be divided in two categories: near-term climate forcers, and well-mixed GHG. Near-term climate forcers include for example black carbon or nitrogen oxides. Near-term climate forcers have very short lifetimes in the atmosphere: days to weeks. Their impact on climate change is dependent on the region of emission and their long-term climate change is negligible. They are currently excluded from the impact characterisation on climate change due to high uncertainty. Well-mixed GHG includes for instance CO_2 and CH_4. Due to their lifetime of several years, decades, or centuries, the impact of well-mixed GHG is independent of the region of emission. In this thesis GHG will refer to well-mixed GHG.

The impact pathway, or cause-effect chain, of climate change begins with a modification of the atmospheric concentration of GHG, due to the GHG emissions (recorded in the LCI). This change in atmospheric concentration leads to a change in radiative forcing. Global warming Potential (GWP) introduced in 1990 in the first assessment report of the IPCC enables to convert emissions of GHG into impact on radiative forcing (UNEP-SETAC 2016). A change in radiative forcing results in a change in atmospheric temperature. The Global Temperature Change (GTP) proposed by Shine et al. (2015) allows evaluating the increase in global temperature compared to the pre-industrial temperature due to GHG emissions. Unlike the GWP, which is a cumulative indicator, the GTP is instantaneous. The change in atmospheric temperature then leads to a variety of impacts (cf. section 1), and thus indicators, for instance, the global sea-level rise potential modelled by Sterner et al. (2014) or the global precipitation change potential modelled by Shine et al. (2015).

Such characterisation methods positioned at the end of the impact pathway decrease uncertainty due to unfair interpretation of the results (UNEP-SETAC 2019) as the results are directly linked to relevant environmental issues. However, the models are more complex, increasing uncertainties on the characterisation factors. Kirschbaum's work (2014) provides a good illustration of the problem of interpreting results obtained with indicators close to the beginning of the climate change cause-effect chain. Kirschbaum (2014) proposes three indicators to assess three aspects of the impact of GHG emissions based on temperature change:

- the amplitude of the temperature change (the greater the amplitude, the greater the risk of droughts and extreme weather events),
- the speed of the temperature change (the faster it is, the less time the environment has to adapt (fauna and flora migration) and the greater the risk of species extinction),
- the duration of the temperature change (the longer the environment is exposed to high temperatures, the more we will be confronted with melting ice and rising water, i.e. reversible events on a longer time scale than the temperature rise).

Jolliet et al. (2018) indicate that the use of GTPs or GWPs alone does not allow for the assessment of both the long-term and short-term effects of temperature changes for a static assessment. Jolliet et al. (2018) thus advise calculating both i) GWP for a time horizon of 100 years, which is a good indicator of short-term effects because these numerical values are close to GTP for a time horizon of 40 years, and ii) the GTP for a time horizon of 100 years for long-term effects. This reveals the complexity of a comprehensive impact assessment in the "climate change" category with a single value.

Currently, the impact of GHG emissions on global warming is often measured using only GWP for time-horizon of 100 years provided and regularly updated by the IPCC (2021). The GWP of a GHG i is calculated according to the following formula (IPCC 2013):

$$GWP_i = \frac{\int_0^{TH} a_i C_i(t)dt}{\int_0^{TH} a_{CO2} C_{CO2}(t)dt} \tag{2}$$

with:

- a_i the instantaneous radiative forcing of the GHG$_i$ which depends on its concentration in the atmosphere. For CO_2, a_i can be considered equal to 5.35 W/m^2 divided by the amount of CO_2 in the atmosphere (kg) for small variations of this concentration (Joos et al. 2013).
- $C_i(t)$ is the function that represents the atmospheric degradation or decay of the GHG$_i$ in the atmosphere over time after its emission.

The parameters of the decay function of CO_2 provided in the latest IPCC report (IPCC 2021) were calculated by Joos et al. (2013). To formulate the decay function of CO_2, they fitted the results of a carbon cycle-climate model run with an emission pulse of 100 GtC, corresponding to 47.10 ppm, and a constant background concentration of 389 ppm (2010 value). 100 GtC is considered as a small perturbation, notably for the calculation of the radiative efficiency. Joos et al. (2013) confirmed the relevance of the calculated GWP to model the impact of "infinitely small carbon addition or removal to the atmosphere", i.e. less than 1 GtC.

Emissions of CO_2 initially captured from the atmosphere, by biomass growth, for instance, are assumed to have no impact on global warming, i.e. a GWP of zero. This assumption is used in national emission inventories to avoid double counting between the land use, land-use change and forestry sector and other sectors, such as energy (Cowie et al. 2021). The use of this assumption in LCA is discussed in the following subsection.

4.2 MOVING AWAY FROM THE ASSUMPTION OF ATMOSPHERIC CO2 NEUTRALITY ON CLIMATE CHANGE

The decay function of CO_2 is calculated using climate models. For instance, Cherubini et al. (2011) used the climate model Bern 2.5CC. This model includes CO_2 capture by the oceans and vegetation. The capture of CO_2 by vegetation is only due to the additional growth of biomass because of the increase in the level of CO_2 in the atmosphere and the increase in fertilisation efficiency. Moving away from the CO_2 neutrality assumption and accounting for CO_2 sequestration by biomass in the context of bioenergy does not therefore lead to double counting. Assuming a symmetry of response of the climate system to uptake and release of CO_2, uptake and release of atmospheric CO_2 can be characterised using the same characterisation factor as fossil CO_2. This approach is referred to as the "+1/-1" approach in the literature (Cucurachi et al. 2022).

Bioenergy was developed with the objective of reducing the climate change impact of the energy production in transport, residential-tertiary or industrial sectors. The main asset of bioenergy is the assumption of atmospheric CO_2 neutrality on climate change. Searchinger (2010) argue that without bioenergy, CO_2 would have still been captured by plants and thus that the CO_2 emitted by bioenergy should not always be counted as neutral. According to Searchinger (2010), only additional carbon, i.e. “carbon that would otherwise be in the atmosphere if not incorporated in biomass used for fuel”, can offset CO_2 emissions of fuel combustion and be considered as impact neutral. Brandão et al. (2013) stressed out that such counting of atmospheric CO_2 may lead to overestimate the impact of CO_2 emissions. In fact, the CO_2 emitted by biomass combustion, additional or not, was initially captured by photosynthesis during biomass growth. Thus, in the case of “not additional” CO_2, counting only CO_2 emissions and no benefit for the initial CO_2 capture, leads to overestimate the increase in CO_2 atmospheric concentration. To evaluate without bias the mitigation potential of bioenergy, Brandão et al. (2013) propose to keep the biogenic CO_2 climate neutrality assumption, and to add in the inventory all the emissions or captures due to soil organic carbon and land use changes.

Another approach is proposed by Albers et al. (2020). Albers et al. (2020) considered the dynamic of emissions and capture of atmospheric CO_2. They provided a decision tree for choosing the "time perspective" of the LCI, i.e. is the CO_2 captured before or after being emitted. Their approach is focused on forest management but could be generalised to any system using biomass. Their proposals can be interpreted relative to the concept of “additional carbon”. The case when the harvested wood comes from a managed forest correspond to additional carbon and an historical perspective must be used. The case when harvested wood does not come from a managed forest correspond to non-additional carbon. If the forest is allowed to regrow, benefice for capturing atmospheric carbon is included with a future perspective. If the forest is not allowed to regrow, no benefice for capturing atmospheric carbon is included. This approach is the most transparent for taking into account the impact of uptake and release of atmospheric CO_2. It requires the use of the “+1/-1” approach and dynamic life cycle impact assessment. Dynamic LCIA is presented in the next section.

4.3 DYNAMIC LCIA

The time dimension appears at two levels in an LCA: the description of the system, and the characterisation of the impact (Beloin-Saint-Pierre et al. 2020). For some inputs, it may be interesting to include their temporal variations in the construction of the LCI. For instance, it is interesting to match, on an hourly basis, the variations in the electricity mix and the electricity consumption by the system, rather than using an average composition over the year. Specific characterisation factors are applied depending on the date of emission. Equation (1) then becomes:

$$I(TH) = \sum_{t} \sum_{s} g_{t,s} CF_s(TH - t) \tag{3}$$

with $g_{t,s}$ the amount of substance s emitted/consumed at time t. This implies building an inventory that contains the necessary temporal information to be able to match inventory data with characterisation factors.

The development of dynamic characterisation factors currently focuses on only three impact categories: climate change, ecotoxicity and ozone depletion (Beloin-Saint-Pierre et al. 2020). Dynamic impact characterisation on climate change is an active field of research with new characterisation methods (based on GWP (Ventura 2022)) and indicators to help decision-making (based on GTP (Tiruta-Barna 2021)) that keep being developed. A summary of existing dynamic indicators to evaluate climate change based on GWP or GTP is provided in Table 2. In dynamic LCA, the impact is calculated using the formulas and not the values of GWP or GTP provided by the IPCC for fixed time horizons of 20, 100 or 500 years. This means that the results can be presented as the evolution of the impact as a function of time horizons. This presentation of results using a curve rather than a single value makes it possible to visualise the short- and long-term impacts. When comparing systems, it is then possible to visualise temporal burden-shifting due to the replacement of short-lived GHG with long-lived GHG, or vice versa, as illustrated in Figure 2.

Table 2: Summary of the existing dynamic indicators to evaluate climate change based on GWP or GTP. i: GHG (ex: CO_2, CH_4), a_i : instantaneous radiative forcing of the GHG_i, $C_i(t)$ decay function of the GHG_i, TH: Time Horizon of the impact assessment. te: time of emission.

Dynamic indicators based on Global Warming Potential		
Formula for an emission peak of i at time t_e	Description	Source
$GWP_i = \frac{\int_0^{TH} a_i C_i(t)dt}{\int_0^{TH} a_{CO2} C_{CO2}(t)dt}$	Recall of the static formula.	(Olivié and Peters 2013)
$Absolute\ GWP_i(t_e) = \int_0^{TH-t_e} a_i C_i(t)dt$	Original dynamic LCA method developed by Levasseur et al. in 2010. The impact of the emissions occurring after the chosen TH are cut-offed.	(Levasseur et al. 2010)
$GWP_i(t_e) = \frac{\int_0^{TH} a_i C_i(t)dt}{\int_0^{TH+t_e} a_{CO2} C_{CO2}(t)dt}$	New proposal in order to comply with two of the principles Ventura sets out and which she considers fundamental for LCA: using the same impact	(Ventura 2022)

	integration period for all substances and all moments of emissions.	
$CTP_i(t_e) = \frac{\frac{\int_{t_e}^{TH} a_i C_i(t)dt}{A_{CO2,ppm}}}{\int_{t_e}^{TH}\left(BC_i(TH) - BC_i(t)\right)dt}$	This indicator evaluates the distance to Climate Tipping Points (CTP). The denominator represents the remaining atmospheric capacity until the target time TH. $A_{CO2,ppm}$ is 'the specific radiative forcing of CO_2 for 1 ppm with a background concentration of 378 ppm'. $BC_i(t)$ is the atmospheric concentration of the GHG i at time t.	(Jørgensen et al. 2014)
Dynamic indicator based on Global Temperature Change		
Formula for an emission peak of i at time t_e	Description	Source
$GTP_i = \frac{\int_0^{TH} a_i C_i(t) IRFT(TH-t)dt}{\int_0^{TH} a_{CO2} C_{CO2}(t) IRFT(TH-t)dt}$	Recall of the static formula. $IRFT$: function representing the 'evolution of the global-mean temperature in response to a radiative forcing'	(Olivié and Peters 2013)
$Absolute\ GTP_i = \int_{t_e}^{TH} a_i C_i(t) IRFT(TH-t)dt$	Dynamic GTP proposed by Shimako et al. in 2018.	(Shimako et al. 2018)

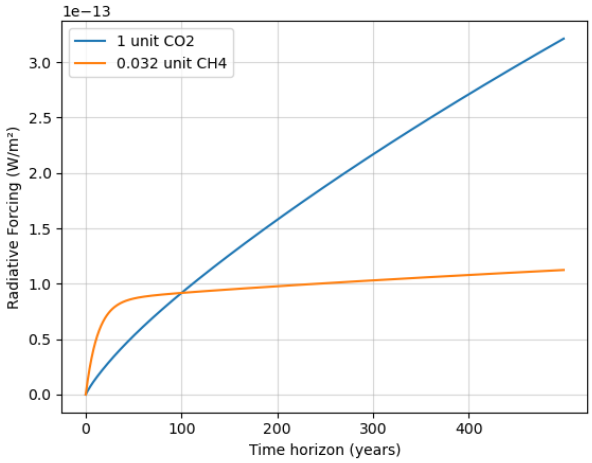

Figure 2: Evolution of the radiative forcing caused by a pulse emission of one unit of CO_2 compared to an emission of CH_4 causing an equivalent impact at 100 years (0.032 unit of CH_4). Such a representation of the impact allows visualising temporal burden-shifting. Replacing an emission of CO_2 by an emission of CH_4 causing an equivalent impact at 100 years would cause temporal burden-shifting: a decreasing impact in the long-term but an increased rate of warming in the short-term.

The impact can be calculated using the absolute GWP (AGWP) without the normalisation of the impact by the impact of a pulse emission of a unit of CO_2. The normalisation of the impact is inspired by the definition of ozone depletion potential and justified as “for simplicity” in the first assessment report of the IPCC (Shine et al. 1990). The interpretation of GWP according to the IPCC is that the GWP represent how much more energy could be avoided over a given time period by avoiding the pulse emission of a unit of GHG compared to avoiding a pulse emission of a unit of CO_2 (IPCC 2021). Nowadays, society is familiar with the unit CO2eq. This has the advantage of facilitating the dissemination of results (UNEP-SETAC 2016). Another advantage raised in the GLAM is the reducing of uncertainty compared to an absolute metric. However, the complexity of the model used to calculate the decay function of CO_2 and its associated uncertainties are still included in the GWP of GHG other than CO_2 (Levasseur et al. 2016; Reisinger et al. 2011). As recalled by the IPCC (2021), emissions metrics “do not define policy goals or targets but can support the evaluation and implementation of choices with multi-component policies”. Thus, policy goals or targets could also be expressed in W/m^2, the unit of radiative forcing. For instance, the planetary boundary for climate is defined using two control variables: “an atmospheric CO_2 concentration of 350 parts per million (ppm) and an increase in top-of-atmosphere radiative forcing of +1.0 W/m^2 relative to preindustrial levels” (Steffen et al. 2015). Therefore, normalising the impact in the impact category “climate change” by the impact of a pulse emissions of a unit of CO_2 is not essential for interpreting results. Moreover, in the first assessment report of the IPCC, GWP is introduced to answer the question “which gases are the most important?” (Shine et al. 1990). This question can be answered in an LCA study by looking at the contribution of each gas to the total impact of the system without the need for the normalisation of the impact by the impact of a pulse emission of a unit of CO_2.

5 RESEARCH QUESTIONS AND STRATEGY

In the previous sections, the necessity of achieving negative emissions to reach carbon neutrality was demonstrated. However, storing CO_2 in geological formations does not create value, and the chemical industry requires carbon-containing raw materials. Thus, CCU is considered an opportunity to generate both value and negative emissions. The environmental relevance of such systems needs to be assessed using LCA, a mature method that still raises methodological questions. This brings us to the research question of this thesis: How to address the methodological challenges associated with the LCA of CCUNET systems?

Firstly, to evaluate negative emissions potential, all the lifecycle steps need to be included in the system boundaries, i.e. “from-cradle-to-grave”, from CO_2 capture from the atmosphere to its permanent storage out of the atmosphere or its release (Tanzer and Ramírez 2019). To facilitate the use of these system boundaries, inventory data must be available for stages that are not the main focus of the LCA study. For CCUNET systems, these will probably be the upstream CO_2 production and downstream end-of-life stages. BECCS systems are the most probable source of atmospheric CO_2 in the near future. LCI data quality is key to the reliability of LCA results. This leads to the question explored in the first article:

What LCI data are used for the LCA of BECCS?

Secondly, if ISO standard (2006a; 2006b) can be applied to every kind of product or service, it has been further specified by government agencies to provide more guidance, with guides such as the Product Environmental Footprint (PEF)(European Commission 2021) or the Publicly Available Specification (PAS)(PAS 2050:2011). Some methodological choices (allocation methods, inclusion or not of life cycle stages, etc.) can cause discrepancies between results of LCAs of similar products. To harmonise LCA results and improve results comparability between studies, LCA guidelines exist for groups of products, such as CCU systems (Ramirez Ramirez et al. 2020; Zimmermann et al. 2020), NET (Goglio et al. 2020) or plastics (Nessi et al. 2021; Nessi et al. 2022). However, neither the LCA guidelines on CCU nor the LCA guidelines on NET contain comprehensive recommendations for conducting an LCA on a CCUNET system. Actually, carrying out an LCA on a CCUNET system generates specific issues, for example, on the choice of the method to solve multifunctionality. This leads to the question answered in the second article:

Are the LCA guidelines on CCU compatible with the evaluation of negative emissions?

Thirdly, the issue of the timing of CO_2 emissions and captures is critical not only for NETs but also for CCU systems (Bui et al. 2018; Goglio et al. 2020). Dynamic LCA offers two major advantages. Firstly, it provides a transparent and more accurate accounting of the impact of uptake and release of atmospheric CO_2 (see section 4.2). Secondly, it offers the possibility of representing the impact on climate change over several time horizons, enabling both short- and long-term impacts to be visualised (cf. section 4.3). Dynamic LCA is not yet included in conventional LCA software. It is not yet very practical to use. This leads to the question answered in the third and final article:

How can we facilitate dynamic LCA?

The aim of this thesis is therefore to contribute to the methodological development of LCA for a better assessment of negative impacts on climate change, focusing on CCU technologies. The research strategy is as follows:

1. Chapter 1: Provide inventory data to be able to use « from-cradle-to-grave » system boundaries

2. Chapter 2: Explore the compatibility of LCA guidelines for CCU systems with the evaluation of negative emissions
3. Chapter 3: Facilitate dynamic LCA to be able to account for the timing of CO_2 uptake and release
4. Chapter 4: Propose a method to evaluate if further investigation using dynamic LCIA is necessary, using only simplified temporal information for enabling LCA practitioners to target their efforts more effectively between improving the quality of inventory data and performing dynamic LCIA

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Chapter 1: Life cycle assessment of bioenergy with carbon capture and storage system: Critical review of life cycle inventories

This first chapter is linked to the methodological challenge of using "from-cradle-to-grave" system boundaries to carry out the evaluation of negative emissions, as explained in the chapter context. This chapter corresponds to the published article: Duval-Dachary S, Beauchet S, Lorne D, Salou T, Helias A, Pastor A (2023) Life cycle assessment of bioenergy with carbon capture and storage systems: Critical review of life cycle inventories. Renewable and Sustainable Energy Reviews 183:113415. https://doi.org/10.1016/j.rser.2023.113415. The purpose is to review the life cycle inventory data that are used for BECCS LCA. To this end, 35 recent BECCS LCA are selected and the inventory data they used is collected. Synthesis inventory tables are then compiled, including the observed range of variability for each data item. Data quality is also reviewed, through the representativeness, reproducibility and completeness of the life cycle inventories highlighted in the selected articles.

List of abbreviations

BECCS	Bioenergy with Carbon Capture and Storage
CHP	Combined Heat and Power
DM	Dry Matter
EOR	Enhance Oil Recovery
FT	Fischer-Tropsch
GHG	GreenHouse Gases
HTT	HydroThermal Treatment
IPCC	Intergovernmental Panel on Climate Change
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LHV	Lower Heating Value
LUC	Land Use Change
MDEA	MethylDiEthanolAmine
MEA	MonoEthanolAmine
MSW	Municipal Solid Waste
NET	Negative Emissions Technologies
N.S.	Not Specified
PVSA	Pressure Vacuum Swing Adsorption
SOC	Soil Organic Carbon
SM	Supplementary Materials
vol%	Percentage based on volume
WGS	Water Gas Shift
WM	Wet Matter
wt%	Percentage based on weight

CHAPTER CONTEXT

Negative emissions are generated by a system when the impact of atmospheric CO_2 uptake and permanent storage is not offset by atmospheric CO_2 release and other GHG emissions during the system entire life cycle. This implies that the evaluation of negative emissions potential requires "from-cradle-to-grave" system boundaries, i.e. from CO_2 capture from the atmosphere to its permanent storage out of the atmosphere or its release, as pointed out by Tanzer and Ramirez (2019). They warn against the misinterpretation and miscount of negative emissions generated by the use of other system boundaries. This could lead to "policy incentives that reward increasing atmospheric greenhouse gas concentrations under the guise of negative emissions" .

Thonemann (2020) reviewed LCA of CCU systems. He observed that 25% of the 44 articles he reviewed did not even specify the source of CO_2 and 80% used system boundaries excluding the end-of-life of the CO_2-based product. Using "from-cradle-to-grave" system boundaries is thus not common is LCA of CCU systems, which hinders the identification of CCUNET systems. CCUNET systems are complex and involve multiple actors (CO_2 source, CO_2 valorisation, CO_2-based product end-of-life and so on) as illustrated in section 3 of the introduction. Due to the complexity of CCUNET system, generic LCI data are necessary to model lifecycle steps that will not be the main focus of future LCA study, such as the CO_2 source or the end-of-life of the product, and use "from-cradle-to-grave" system boundaries.

Concerning the CO_2 source, Thonemann (2020) observed that 65% of the 44 articles choose a BECCS system as CO_2 source (biogas upgrading to biomethane, combustion or fermentation). As explained in the introduction (see section 2), BECCS systems are included in the European and French strategy to mitigate climate change, along with DACCS. However, DACCS is a less mature technology than BECCS systems, with notably a much higher cost. BECCS systems are thus the most probable source of atmospheric CO_2 in the near future. Concerning the end-of-life of the CO_2-based product, BECCS systems include incineration with CCS, one of the end-of-life options with the potential to generate negative emissions. The focus of this chapter is thus BECCS systems.

As explained in the introduction, LCI data quality is key to the reliability of LCA results. Therefore, the aim is to review the recently published articles on LCA of BECCS to compile generic LCI tables, containing values range for uncertainty analysis, for the main lifecycle steps of a BECCS system. The quality of the data used is also analysed to check if the current evaluation of BECCS system is based on robust data.

1 INTRODUCTION

In 2018, the United Nations declared that climate change affected the lives of 39 million people, due to extreme weather events, changing weather patterns and sea-level rise (United Nations 2019). According to the latest report of the Intergovernmental Panel on Climate Change (IPCC 2022), an increase in global temperature will result in further degradation of human health and ecosystems, in rising food and water insecurity and in the destruction of infrastructure. In 2019, the planetary boundary for climate change (uncertainty range of 350 to 450 ppm (Steffen 2015)), was reached, with a CO_2 concentration in the atmosphere of 409.9 ppm. Taking “urgent action to combat climate change and its impacts” is thus the thirteenth sustainable development goal defined by the United Nations (2019). To take action, 196 nations signed the Paris agreements. The Paris agreements aim at limiting global warming to 2°C, or even 1.5°C (UNFCCC 2020). Starting from 1st January 2020, the remaining carbon budget to limit global warming to 1.5°C is estimated to be around 500 $GtCO_2$ (Arias et al. 2021). Given the current rate of CO_2 emissions, i.e. 33 Gt/year (IEA 2021), and their historical continuous increase, this budget will be spent in less than 15 years. Humanity therefore needs to urgently achieve net zero CO_2 emissions in order to combat climate change.

Mitigation measures include the reduction of energy and material consumption (sufficiency and efficiency), using decarbonized technologies and carbon dioxide removal, also known as negative emission technologies (NET) (IPCC 2018). NET are defined as “intentional human efforts to remove CO_2 emissions from the atmosphere” (Minx et al. 2018). If the remaining carbon budget were to be exceeded, NET could also be used to compensate for this overshoot. The IPCC points out that relying only on NETs to limit global warming is reckless (IPCC 2018). Nevertheless, across scenarios limiting global warming to 1.5°C, the median for bioenergy with carbon capture and storage (BECCS) deployment range from 3 to 7 $GtCO_2$ per year by 2050 (IPCC 2018). BECCS also produce renewable energy, making it an attractive and widely studied NET. The definition of BECCS, sometimes also abbreviated as Bio-CCS, slightly differs throughout the literature as was highlighted in the work of Kemper (2015). In the present work, BECCS systems are divided into four steps: i) B, biomass production, ii) E, conversion of biomass into energy, in all its forms (electricity, heat and transportation fuels), iii) CC, capture of the produced CO_2 (pure or diluted in flue gas or syngas) and iv) S, CO_2 transport and storage. A wide diversity of BECCS systems exist due to the numerous technological options available for each of the four life cycle steps. For instance, the existing processes to convert biomass into energy (combustion, gasification…) are described in the review of Shahbaz et al. (2021). The CO_2 capture processes (post-combustion, pre-combustion, oxy-fuel) are presented in the IPCC special report on Carbon Dioxide Capture and Storage (IPCC 2005) or more recently in the review of Raynal and Tebianian (2020). All BECCS systems have the potential to produce negative emissions. Indeed, BECCS systems intentionally remove CO_2 from the atmosphere by photosynthesis during biomass growth (step i), capture it during the combustion of the biomass (step ii and iii) and store it over long-term in geological formations (step iv). Each BECCS system faces multiple challenges (Bui et al. 2018; Creutzig 2015; Kemper 2015; Shahbaz et al. 2021): sustainability and availability of biomass, public perception, policy, regulatory, technical and economic issues. Decision-makers need to be able to select the optimal BECCS systems to deploy, in order to follow the IPCC 1.5°C or 2°C scenarios.

To support this decision, the Life Cycle Assessment (LCA) of each BECCS system is required. LCA is a standardised and recognised method for performing the environmental assessment of products and services (ISO 2006). An environmental evaluation of a BECCS system from a life cycle perspective is necessary to calculate the true negative emissions achieved. Achieving true negative emissions requires two conditions to be fulfilled: i) greenhouse gases (GHG) must be removed from the atmosphere and stored; ii) the GHG emissions over the whole life cycle of the system (from biomass growth to CO_2 storage) must not offset the amount of GHG removed (Tanzer and Ramirez 2019). Indeed, the production of energy and chemicals that are consumed during processes (e.g. CO_2 capture) leads to additional emissions of greenhouse gases. BECCS may also be responsible for other types of environmental impacts, such as acidification, water footprint or human toxicity (Oreggioni et al. 2017). LCA addresses these potential trade-offs between climate change and other impact categories. Goglio et al. (2020) have already proposed recommendations to carry out LCA evaluations of NET. Furthermore, the methodological issues (allocation method or functional unit definition) linked to the application of LCA to NET systems have been reviewed by Creutzig et al. (2015), Goglio et al. (2020) and Terlouw et al. (2021). The impact on climate change of BECCS processes have been reviewed by Shahbaz et al. (2021) and Li et al. (2020). The reviewed magnitude of negative emissions generated by BECCS is variable and can sometimes even be positive (Li and Wright 2020; Shahbaz et al. 2021; Terlouw et al. 2021). Overestimating the environmental benefit of BECCS would imply deviating from climate change mitigation trajectories. It is thus important to discuss the causes for variability in results and the quality of the conducted LCAs. In Li et al. (2020), the reported impacts on climate change of different types of BECCS range from -1.5 $kgCO_2eq/MJ$ to 0.4 $kgCO_2eq/MJ$. According to Terlouw et al. (2021), this variability arises from methodological choices and modelling assumptions (ex: system boundaries or allocation).

As for the methodological choices, the choices made for building a Life Cycle Inventory (LCI) can also be a source of result variability. An LCI assembles all emissions to the environment as well as the resource consumption from the environment caused by the system under study. Collecting data to build the LCI is an essential but time-consuming part of conducting an LCA. During data collection, LCA practitioners are confronted with a variety of choices, for example, concerning the method to use in order to fill in missing data. Sometimes LCA practitioners can also have no other choice but to use the only available data. Knowledge on the quality of the data relatively to the purpose of the LCA provides the reader with adequate information on the relevance of the results obtained. Moreover, an assessment of the variability of LCI data allows for the uncertainty of the results to be calculated. Thus, the relevance of the LCA results strongly depends on the LCI data quality. According to the LCA standard (ISO 2006), data quality encompasses temporal factors, geography, technology, fidelity, completeness, representativeness, consistency, reproducibility, data source and information uncertainty (see section 2 for the definitions). To follow the LCA standard (ISO 2006), an LCA practitioner should thus make sure that the quality of each LCI data item is consistent with the goal initially defined. Depending on the goal, default inventories can also be useful and relevant to model some of the life cycle steps. Performing an LCA on BECCS has various goals: for instance, to identify hotspots in processes under development, to evaluate deployment scenarios or to compare CO_2 capture processes. However, the system boundaries should systematically include all the steps from biomass growth to CO_2 storage in order to enable comparison among studies. Unfortunately, generic LCI are not yet available at present for every life cycle steps. For example, the worldwide commonly used Ecoinvent LCA database v3.8. does not include any CO_2 capture processes.

This highlights the main research question of this study: what LCI data is used for the life cycle assessment of BECCS? To answer, a review of recent articles on LCA of BECCS systems was carried out following the methodology presented in section 2. Resulting from this review, a state-of-the-art can be established on the previously evaluated BECCS systems, using the LCA methodology presented in section 3.1. The LCI inventory datasets built from data obtained from these articles are provided in section 3.2. These datasets include a range of variations for most inputs in order to facilitate uncertainty studies. The quality of the data provided in the articles is synthesised in section 3.3, then discussed in section 4.1. Lastly, section 4.2 highlights the BECCS systems that still need to be evaluated from a life cycle perspective.

2 MATERIAL AND METHODS

Technologies in the CCS sector are developing at a rapid pace. Focus was therefore solely put on peer-reviewed articles dating between 2015 and 2021. To identify scientific articles on bioenergy, LCA and CCS, a database literature review was conducted using SCOPUS (field = “Article title, Abstract, Keywords”) and Web of science databases (field = “all Fields”) with the following keywords on November 2021 (U in BECCUS stands for Utilisation):

- Bioenergy: "biofuel" or "bioenergy" or "BECCS" or "BECCUS" or "BECCU" or "biochar"

- Life cycle assessment: "LCA" or "Life cycle assessment" or "Life cycle analysis" or "Life cycle impact assessment”
- Carbon capture and storage: "CCS" or "Carbon capture and storage" or "CO_2 capture and storage"

97 articles were identified. The eligibility criterion was that a LCA on at least one BECCS system was performed in the article. After screening by one reviewer, only 35 precisely covered LCA on BECCS and were thus included in this review (see Table 3). Further information on the selection process of the articles is provided in the supplementary materials (SM).

This review focusses on LCI data. In the LCI, the input and output data of each process involved in the life cycle of the product are quantified. Processes are either part of the foreground or of the background systems. The foreground, and the background systems, respectively, “consist of processes which are under [and respectively not or indirectly under] the control of the decision-maker for which an LCA is carried out”(Frischknecht 1998). Background LCI data are usually extracted from databases, such as Ecoinvent. Each background dataset used to model each foreground flow is unfortunately rarely mentioned in the articles. Therefore, this review does not evaluate the representativeness of background datasets. This review rather focuses on foreground data made available in the selected articles. Since there is no consensus on the methodology for assessing data quality in LCA (Edelen and Ingwersen 2018), the aspects of data quality studied in this review are based on data quality requirements as defined in ISO 14044 (2006). The considered data quality criteria and corresponding collected data items are indicated in Figure 3.

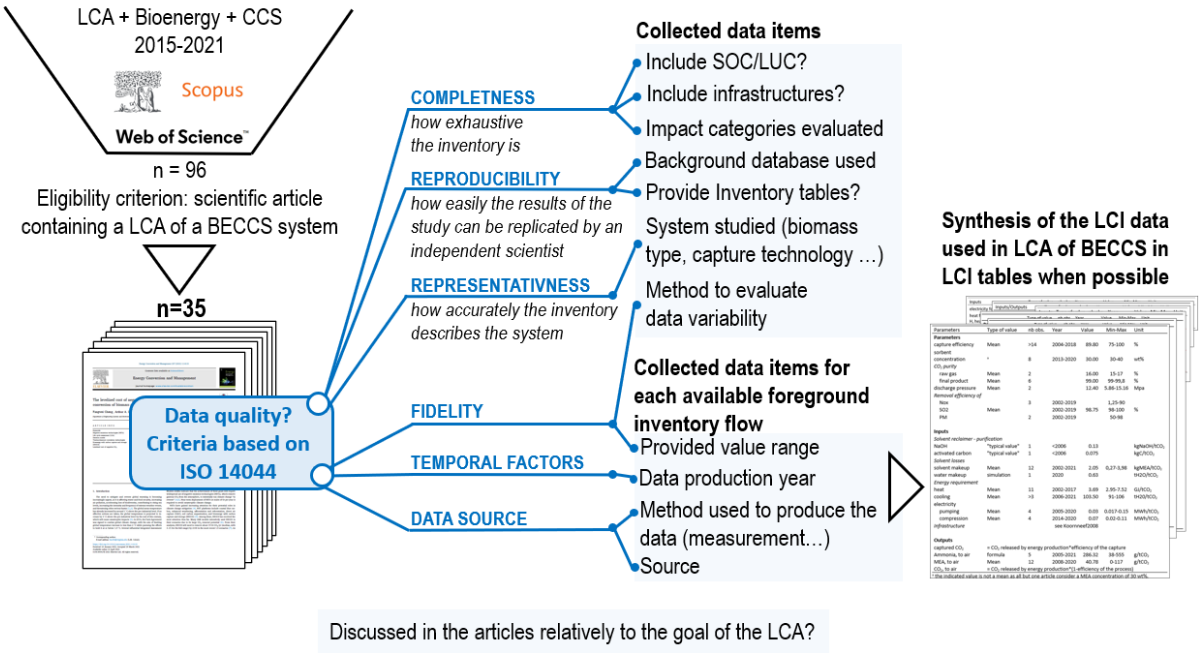

Figure 3: Method followed to perform the review. BECCS: Bioenergy with Carbon Capture and Storage; CCS: Carbon Capture and Storage, LCA: Life Cycle Assessment; SOC: Soil Organic Carbon; LUC: Land Use Change

The criterion "consistency" is defined in ISO 14044 (2006) as the "qualitative assessment of how the study methodology is applied consistently to the different components of the analysis" (author's translation). This "consistency" criterion lies outside the scope of the research question of this review. The "uncertainty" criterion is approached through the "fidelity" criterion.

Thus, for each foreground LCI flow, four types of information were collected: the method of production, the source, the year of production, and the value range. Based on this inventory (available in the SM), synthesis tables of foreground processes are built when a sufficient amount of data is available. The values indicated in these tables either correspond to the value provided in an article if its quality is higher than for the other sources (far more recent or value from industrial source, indicated as "IV" (Industrial Value)) or to a mean value (indicated as "mean").

The SankeyMATIC software (Bogart 2015) was used to produce the following Sankey diagrams. Further information is given in the SM.

3 RESULTS

In this section focusing on results, the numerical value and quality of the selected BECCS LCIs are studied in order to analyse the data used to perform BECCS LCA. To this end, the results are divided in three steps. First, the selected articles on which this review is based are presented through i) Table 1 and ii) an overview of the BECCS systems they evaluated. Second, the necessary elements to support the discussion on data quality (reproducibility, completeness, data source of the provided LCI data and data production date) are reviewed. Third, site-generic inventory tables are compiled in order to facilitate future assessments of BECCS in a consistent manner. These tables include biomass harvesting and pretreatment, combustion, gasification, monoethanolamine (MEA) capture, pipeline transport and geological storage.

Table 3: List of the selected articles and pieces of information about data quality. CC: Climate Change; SOC: Soil Organic Carbon; LUC: Land Use Change; n.s.: not specified, b.o.: for the biomass production step only

Authors	Year	Data variability ?	Database used	Infrastructure?	SOC/LUC?	Only CC?	Inventory tables?
Wu and Zhai	2021	sensitivity analysis	Ecoinvent	yes	no	no	no
Cheng et al.	2021	n.s.	GREET	n.s.	n.s.	yes	yes
Kar et al.	2021	n.s.	n.s.	no	yes	yes	no
Wang et al.	2021	n.s.	n.s.	n.s.	n.s.	yes	no
Garcia-Freites et al.	2021	n.s.	Ecoinvent v3.4	yes	no	yes	b.o.
Bennett et al.	2021	sensitivity analysis	GREET 2019	no	n.s.	yes	no
Yang et al.	2021	n.s.	n.s.	yes	yes	yes	no
Bressanin et al.	2021	sensitivity analysis	Ecoinvent v3.4	n.s.	n.s.	no	no
Yan et al.	2021	n.s.	-	no	n.s.	no	yes
Antonini et al.	2021	sensitivity analysis	Ecoinvent v3.5	yes	n.s.	no	yes
Valente et al.	2021	sensitivity analysis	Ecoinvent v3.5	n.s.	yes	no	no
Mohamed et al.	2021	sensitivity analysis	n.s.	n.s.	n.s.	yes	yes
Sproul et al.	2020	n.s.	n.s.	yes	n.s.	yes	no
Bello et al.	2020	sensitivity analysis	Ecoinvent v3.5	no	n.s.	no	yes
Melara et al.	2020	Monte Carlo	Ecoinvent v3	no	no	yes	no
Yang et al.	2020	sensitivity analysis	Ecoinvent, GREET, U.S. LCI database	n.s.	n.s.	yes	b.o.
Field et al.	2020	n.s.	GREET 2018 database v13395	n.s.	yes	yes	no
Hammar and Levihn	2020	sensitivity analysis	Ecoinvent v3	n.s.	no	yes	no
Antonini. et al.	2020	n.s.	Ecoinvent v3.5	yes	yes	no	yes
Zang. et al.	2020	Monte Carlo	ELCD v3.2	yes	n.s.	no	no
Gelfand. et al.	2020	n.s.	GREET 2017	n.s.	yes	yes	no
Cheng et al.	2020	n.s.	GREET	n.s.	yes	yes	yes
Lask et al.	2020	sensitivity analysis	Ecoinvent v3.5	no	yes	no	b.o.
Bennett et al.	2019	Monte Carlo	Ecoinvent 2013	no	n.s.	no	no
Cumicheo et al.	2019	n.s.	n.s.	n.s.	n.s.	yes	no
Yang et al.	2019	sensitivity analysis	Ecoinvent 2004	no	yes	no	yes
Yi et al.	2018	sensitivity analysis	n.s.	no	n.s.	yes	yes
Cavalett et al.	2018	sensitivity analysis	Ecoinvent v3	yes	no	no	yes
Pour et al.	2018	sensitivity analysis	Ecoinvent v3	yes	no	no	no
Tang and You	2018	sensitivity analysis	Ecoinvent v3.3	yes	no	no	yes
Fajardy and Mac Dowell	2017	sensitivity analysis	n.s.	n.s.	yes	yes	b.o.
Oreggioni et al.	2017	n.s.	Ecoinvent v2.2	yes	n.s.	no	yes
Liu et al.	2017	sensitivity analysis	Ecoinvent v3	n.s.	n.s.	no	yes
Lausselet et al.	2017	n.s.	Ecoinvent v3.2	yes	no	no	no
Jana and De	2016	sensitivity analysis	Ecoinvent v3	yes	no	no	no

3.1 EVALUATION OF BECCS SYSTEMS WITH LCA: STATE-OF-THE-ART

In this review, the life cycle of a BECCS system is divided into four steps: i) biomass production, ii) biomass to energy conversion process, iii) CO_2 capture, iv) CO_2 capture and storage. As shown in Figure 4, multiple options exist for each step. These options are combined to get a complete BECCS system, i.e. a "case study". A total of 109 case studies were identified within the 35 selected articles. Figure 4 summarizes the case studies included in these 35 LCA studies, i.e. it does not represent all the existing possibilities of BECCS. Figure 4 shows that all the BECCS systems studied in the identified LCAs are classified in the advanced category, i.e. using feedstock listed in Part A of the Annex IX of the Renewable Energy Directive (European Parliament and Council 2018). Although not definitive, this list of feedstocks involves biomass that has no direct competition with food or feed and corresponds to different kinds of biomass residues, such as waste, lignocellulosic material and crops, and algae. Biomass pretreatment is not shown on Figure 4 for clarity reasons but includes drying, grinding, pelleting and torrefaction. Torrefaction is only studied by Yang et al. (2019). Figure 4 shows that combustion and gasification are the most studied processes to convert biomass to energy. The most studied process for CO_2 capture is MEA-based capture. CO_2 capture processes are divided into three types of processes: post-combustion, pre-combustion and oxy-fuel. On Figure 1, “amine-based MEA”, “second generation” and “PVSA” are post-combustion processes, i.e. CO_2 is captured in the combustion flue gas. “PVSA” can also be used for pre-combustion capture, i.e. applied to syngas, to separate CO_2 from H_2. The principle of oxy-fuel capture is to use pure oxygen rather than air for the combustion. Thus, the resulting flue gas is almost only composed of CO_2 and water, which are easy to separate. Finally, Figure 1 shows that CO_2 transport and storage are the least described steps of BECCS systems. 22 out of the 35 LCAs indicate that CO_2 is transported by pipeline. Only one article also considered CO_2 ship transportation (Hammar and Levihn 2020).

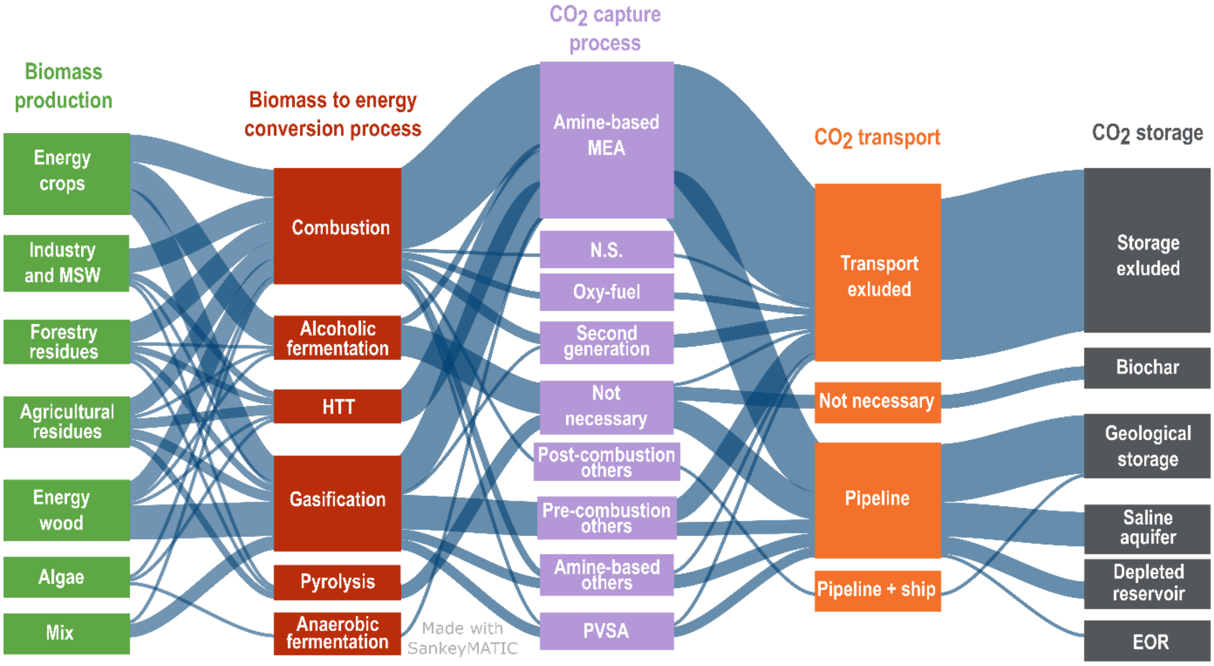

Figure 4: Composition of the case studies identified in the selected articles. The thickness of a flow between two options is proportional to the amount of case studies that include the two options. See Figure 6 and Figure 7 for more details about CO_2 capture processes linked to combustion and gasification respectively. MSW: Municipal Solid Waste; HTT: Hydothermal Treatment; MEA: Monoethanolamine; PVSA: Pressure Vaccum Swing Adsorption; n.s.: not specified; EOR: Enhanced Oil Recovery

3.2 LCI DATA QUALITY

Information on data quality is important to understand the reliability of the results (ISO 2006). Data quality is often reduced to the criteria on “fidelity” and “uncertainty” as defined in the ISO 14044 (2006). Thus, the impact of data variability is assessed in 18 articles using sensitivity analysis, and in three articles using a Monte-Carlos analysis, see Table 3. In any case, the data quality must be evaluated relatively to the goal of the study. The quality of data on each ISO criterion and its relevance to the goal were never entirely discussed or stated in the studied articles.

In the following subsections, only the reproducibility, completeness, and data source of the provided LCI data are reviewed. Data production date and data variability are already indicated in the inventory tables (see previous section and SM for further details). Data production year ranges from 1995 to 2021. Representativeness is discussed in section 4.1.1.

3.2.1 Reproducibility

Only four out of 26 articles using databases (Ecoinvent, GREET (Wang et al. 2021), …) do not indicate the version of the database, see Table 3. 13 articles provide LCI data in the form of LCI data tables for all the life cycle steps, see Table 3. Four provide LCI data tables only for the biomass production step.

The remaining articles either: i) describe the system qualitatively, ii) describe the system partially quantitatively through the text, iii) quote sources, iv) provide simulation results (i.e. raw data), or v) provide only several quantitative parameters in a table. Thus, concerning fermentation, only one article detailed the consumption of chemicals. Some inventory tables are provided after allocations between multiple products. Allocations were used in nine articles (exergy or economic) (see SI). All but one mentioned the allocation factor applied. Among the 13 articles providing LCI tables, only five provide full names (including location) of datasets from the background database used to model foreground input or output flows.

3.2.2 Completeness

The completeness of the inventory depends on the goal of the study. In this review, the completeness is assessed to perform a multicriteria analysis. Very little data were found on infrastructure building and decommissioning while one third of the articles do not even mention infrastructures, see Table 3. Another third of the articles mention infrastructure but do not include it for two reasons: i) the infrastructure was built for a former usage (e.g. gas pipeline) and only reused, or ii) it has a negligible impact on climate change. During biomass storage, there may be other types of consumption of materials (e.g., tarpaulins to protect straw) and energy (e.g., fan to stay dry). For biomass pelleting, no consumption of binder was indicated. For gasification, only three out of 14 publications mentioned bed materials, and only one provide inventory values. For carbon capture and storage, see Table 8. Treatment of the waste stream is also often ignored, such as the treatment of ash from combustion or the decommissioning of the infrastructure. GHG emissions represent the only emission considered in 17 articles, see Table 3. Since one of the functions of a BECCS system is to contribute to climate change mitigation, these 17 articles assess the impact on climate change only. Thus, their inventory was built accordingly. The emissions due to machinery use for harvesting are also not always indicated. The emissions or sequestration of CO_2 due to land use change (LUC) and soil organic carbon (SOC) are discussed in only one third of the articles. Two articles justify their choice for not calculating LUC or SOC: i) for Antonini et al. (2020) the chosen feedstock is not concerned with LUC issues (allocated to the main product), and ii) for Garcia-Freites et al. (2021) the uncertainties on the quantification of SOC are still too high and no standard procedures can be applied.

3.2.3 Data source

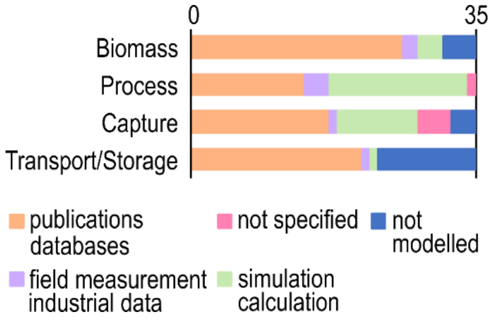

Figure 5: Type of data source to model foreground processes by life cycle step. Process = biomass to energy conversion process

The type of data source used to model foreground processes by life cycle step is illustrated in Figure 5. The source of each collected LCI data is provided in the SI. Biomass production is modelled using data produced either by monitoring, estimations from machinery technical data or taken from the literature. Biomass to energy conversion processes are mainly modelled from simulation of processes (Aspen Plus, IECM) and with data from literature. Simulation of biomass to energy conversion processes often includes simulation of the CO_2 capture process. On the 23 articles that use bibliographic data to model the biomass production steps, 10 use datasets from background databases and 9 use a compilation of literature sources, i.e. two or more sources. In comparison, on the 20 articles that use bibliographic data to model the CO_2 transport and storage steps, 2 use Ecoinvent to model pipelines and only 5 use a compilation of sources. Moreover, three sources are often used: i) Koornneef et al. (2008, quoted in 5 articles) based on the 2006 guidelines for national greenhouse gas inventories of the IPCC and on the work of Damen et al. (2006) (quoted in one more article), and ii) the thesis of Wildbolz (2007, quoted in 4 articles). The work of Rao and Rubin (2002; 2006) was quoted in six publications to model MEA-based capture and is also used by the IECM simulation software. Several options are used for evaluating emissions or sequestration of CO_2 due to LUC and SOC. Cavalett et al. (2018), Valente et al. (2021), Cheng et al. (2020) and Hammar and Levihn (2020) use data from the literature. Yang et al. (2019) and Gelfand et al. (2020) use on site measurements. Lask et al. (2021) follow the recommendations of the ILCD handbook. Yang et al. (2020), Kar et al. (2021) and Field et al. (2020) use models to calculate SOC (DNDC), land use change (CCLUB tool) or both (DayCent) respectively.

3.3 SYNTHESIS TABLES

In the following section, site-generic inventory tables are proposed for biomass harvesting and pretreatment, combustion, gasification, MEA capture, pipeline transport and geological storage. Some processes are not synthesised in the form of an inventory table. There are two reasons for this: i) the excluded process can be site-specific, or ii) available data in the selected publications are too scarce. Hence, on the one hand, the production of biomass can be site-specific: Land clearing for biomass production depends on its previous use; Fertilization and irrigation depend on pedoclimatic conditions and agricultural practices; Storage and transport of biomass depend on logistic choices. One the other hand, pyrolysis and hydrothermal treatment (HTT) with CCS, also called liquefaction, are only studied in three articles. Two of the articles are from the same authors (Cheng et al. 2020; Cheng et al. 2021) and do not show any differences in the LCI data. Anaerobic fermentation with CCS is only studied by Melara et al. (2020), on aquatic biomass. The LCI data collected is still available in the SI of this review. However, they were not sufficient to build relevant synthesis tables.

Each following subsection corresponds to a life cycle stage. The synthesis tables are provided, preceded by a brief description of the process. Complementary information (reference publication, database) is also given when relevant.

3.3.1 Biomass production: harvest and pre-treatment

The analysis of the reviewed LCIs of biomass production highlights three key intrinsic properties of biomasses: i) its carbon content, ii) its lower heating value (LHV) and iii) its moisture content. The carbon content is used for calculating the amount of CO_2 stored by the biomass. The LHV is used for calculating the amount of biomass required to produce a unit of energy. The moisture content is used for calculating the necessary amount of energy for drying the biomass. It is also for conversion between a dry basis (indicated as DM) and a wet basis (indicated as WM). The intrinsic properties of biomass are available in the phyllis2 database. The fraction of biogenic material is an additional property to be considered for municipal solid waste (MSW). In fact, MSW is composed of both biogenic and fossil materials (ex: plastic). Only the CO_2 stored by biogenic materials will lead to negative emissions. As a first approximation, the fraction of biogenic material in MSW can be assumed to be equal to 75% of the total carbon contained in the MSW. (83% in Pour et al. (2018), 61.7 to 77.1% in Lausselet et al. (2017)).

Numerous LCA databases contain data about biomass production: Ecoinvent, Agribalyse (ADEME) for French products, GREET® (Wang et al. 2021) for USA, AusLCI for Australia. In addition, based on a literature review, Fajardy and Dowell (2017) provide averaged fertilization values and yield for wheat straw, miscanthus, switchgrass and short rotation coppice willow in Brazil, China, Europe, India and USA. Once produced, biomass types can be grouped into three categories, which share similar harvesting processes: i) herbaceous crops (ex: miscanthus) and straw, ii) wood (ex: poplar), and iii) forestry residues. Herbaceous crops and straw are first cut, swathed, and then baled for storage. Wood is cut and transported out of the forest (forwarding). Forestry residues are collected and chipped to ease storage and transport. As reflected in Table 4, often the only LCI data provided is the consumption of diesel to fuel harvest machinery. However, in Liu et al. (2017) and Cavalett et al. (2018), the consumption of lubricating oil is indicated (see SM). Oreggioni et al. (2017) also considers the transport of machinery and tire wear. When infrastructures, i.e. machinery, are considered in the LCI of biomass production, they are modelled with dataset from the Ecoinvent database (v2.2, v3 or v3.5).

Table 4: Harvesting - synthesis inventory table

Inputs	Type of value	Nb obs.	Year	Value	Min-Max	Unit
Herbaceous crops/straw						
Diesel	mean	<5	2012-2020	2.04	1.52-2.4[a]	L/t_{WM}
Wood						
Diesel	mean	3	2005-2016	1.87	1.71-2	L/t_{WM}
Residues						
Diesel	mean	2	2005-2016	2.88	1.9-3.9	L/t_{WM}

[a] [3.25 L/t_{WM} – 6.85 L/t_{WM}]: maize harvesting in (Yang et al. 2021)

After harvesting, storage and transport depend on logistic choices related to availability of biomass around the plant. Only the associated loss of organic matter over time due to natural decomposition is addressed in the publications. Thus Yang et al. (2019) estimates the losses due to storage and transport to be 1 weight(wt)% on a wet basis. Cavalett et al. (2018) consider 2 wt% losses on a dry basis during transport. Oreggioni et al. (2017) indicate an “average wood chip degradation of 0.03667 vol%/day” and a storage period of 14 days, resulting in a 0.5 vol% loss on a dry basis.

Pre-treatment depends on the processes chosen to convert biomass into energy. The goal of pre-treatment is to facilitate transport and handling (Shahrukh et al. 2016) and/or to have a sufficiently homogeneous feedstock, both in size and composition, in order to control its flow efficiently (Molino et al. 2016; Haider and Seguin 2012). This implies grinding and drying (see Table 5). Pelleting or pelletizing consists of finely grinding biomass in a mill (<3.2 mm) and then extruding and compressing it into pellets (Shahrukh et al. 2016). A binder is sometimes used but it must represent less than 3 wt% of the pellet (Camia et al. 2021). The value indicated for electricity consumption during pelleting varies widely from one publication to another, even for the same kind of biomass. It ranges between 14 and 827 kWh/t_{DM} (7 values) with an average value of 234 kWh/t_{DM}. Alcoholic fermentation of lignocellulosic biomass demands extensive pretreatment to extract the cellulose (for more details see SM). For gasification, the moisture of the biomass should remain below 25-30 wt% (Molino et al. 2016).

According to the type of gasifier, the biomass may need to be grinded: particle sizes range between 0.1-1 mm for entrained flow reactor, between 20-100 mm for fluidized and fixed bed reactor, while no restriction applies for plasma reactors and rotary kiln reactors (Molino et al. 2016). For biomass combustion, the size limit is 100 mm and humidity is maintained below 50 wt% (Haider and Seguin 2012), the optimal moisture content being 10-15 wt% (Fajardy and Dowell 2017). Drying is not necessary prior to anaerobic digestion and hydrothermal liquefaction.

Table 5: Biomass grinding and drying – synthesis inventory table

Inputs	Type of value	Nb obs.	Year	Value	Min-Max	Unit
Electricity - grinding	mean	4	2000-2013	30.19	24.1-71.2	kWh/t_{DM}
Heat - drying	=mass feedstock*(moisture content wet - moisture content dried)*H					
H, heat to evaporate H$_2$0	thermodynamic calculation			3806	3555-4267	MJ/tH$_2$0 evaporated

3.3.2 Biomass to energy conversion process

Combustion

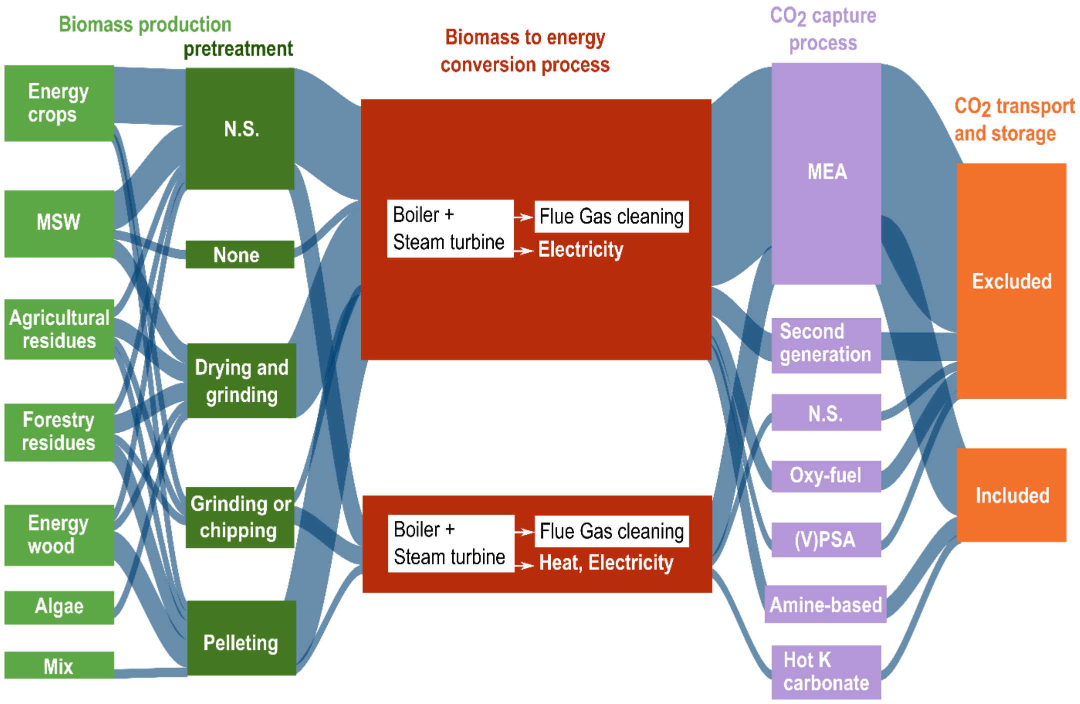

Figure 6: Overview of the systems studied in the 35 LCA of BECCS that include combustion of biomass. The thickness of a flow between two options is proportional to the amount of case studies that include the two options. CHP: Combined Heat and Power; K: potassium; MEA: Monoethanolamine; MSW: Municipal solid waste; n.s.: not specified; (V)PSA: Vaccum Pressure Swing Adsorption

Figure 6 shows that BECCS systems including combustion followed by MEA-based capture have been most referenced (12/35 articles). Values given in Table 6 concern combustion without CCS in the case of a boiler followed by a steam turbine to produce heat and electricity (cf. Figure 6) Information about building and decommissioning of infrastructure is available in the articles of Tang and You (2018) and Koornneef et al. (2008). Other minor flows could be added to fully complete this inventory. In Tang and You (2018), diesel is consumed to maintain a temperature of 850°C in the furnace when the power plant is not producing energy. Oreggioni et al. (2017), Lausselet et al. (2017) and Tang and You (2018) provide supplementary minor emissions. Only electricity can also be produced by the system described in Table 6. The yield is thus a bit higher since no steam is extracted for heat production. It ranges between 32,4%LHV (Wu and Zhai 2021) to 40,5%LHV (Al-Qayim et al. 2015). For the same reason, less water would also be consumed (makeup H20 – steam = 0). A detailed description of the process is presented in the SM.

Table 6: Biomass combustion – CHP (Combined Heat and Power) - synthesis inventory table. kg feedstock as received in the power plant. IV: "Industrial Value"

	Type of value	Nb obs.	Year	Value	Min-Max	Unit
Parameters						
Efficiency						
Electricity - CHP	IV	4	2015	18.00	14-27	%LHV
Heat - CHP	IV	4	2015	63.70	30-74	%LHV
Capacity factor	mean	5	2012-2021	81.85	62.3-91.3	%
Plant lifetime	mean	3	2011-2018	25.00	20-30	yr
Inputs						
Biomass				1.00		kg
Makeup water	mean	3	2019-2021	1.27	0.9-1.5	m3/t feedstock
Water - steam	= output heat [kJ]/enthalpy (2450 [kJ/kg])					
Flue gas treatment						
Electricity	already contained in the value of efficiency of the plant					
Limestone	mean	5	2014-2020	8.43	2.67-14.2	g/kg feedstock
Activated carbon	IV	2	2015	0.45	0.88	g/kg feedstock
Ammonia	mean	4	2014-2021	3.23	0.6-6.6	g/kg feedstock
Water	negligible in front of makeup water					

	Type of value	Nb obs.	Year	Value	Min-Max	Unit
Outputs						
Electricity	=input biomass*LHV biomass*efficiency					
Heat	=input biomass*LHV biomass*efficiency					
Emissions						
CO_2	=C content of the biomass*44/12*input of biomass					
CH_4	literature	1	<2006	0.34	-	g/kg feedstock
N_2O	literature	1	<2006	0.22	-	g/kg feedstock
CO	mean	3	2010-2015	0.25	0.11-0.54	g/kg feedstock
NO_x	mean	4	2013-2019	1.01	0.05-1.88	g/kg feedstock
SO_2/SO_X	mean	4	2013-2019	0.17	0.03-0.5	g/kg feedstock
PM	mean	3	2010-2019	0.09	0.01-0.2	g/kg feedstock
HCl	mean	3	2010-2019	68.69	2.26-190	mg/kg feedstock
NH_3	mean	4	2010-2019	0.05	0.01-0.15	g/kg feedstock
Waste, to be landfilled						
Gypsum	mean	2	2014	1.86	1.29-2.43	g/kg limestone
Ash	=fraction of ash*input biomass					

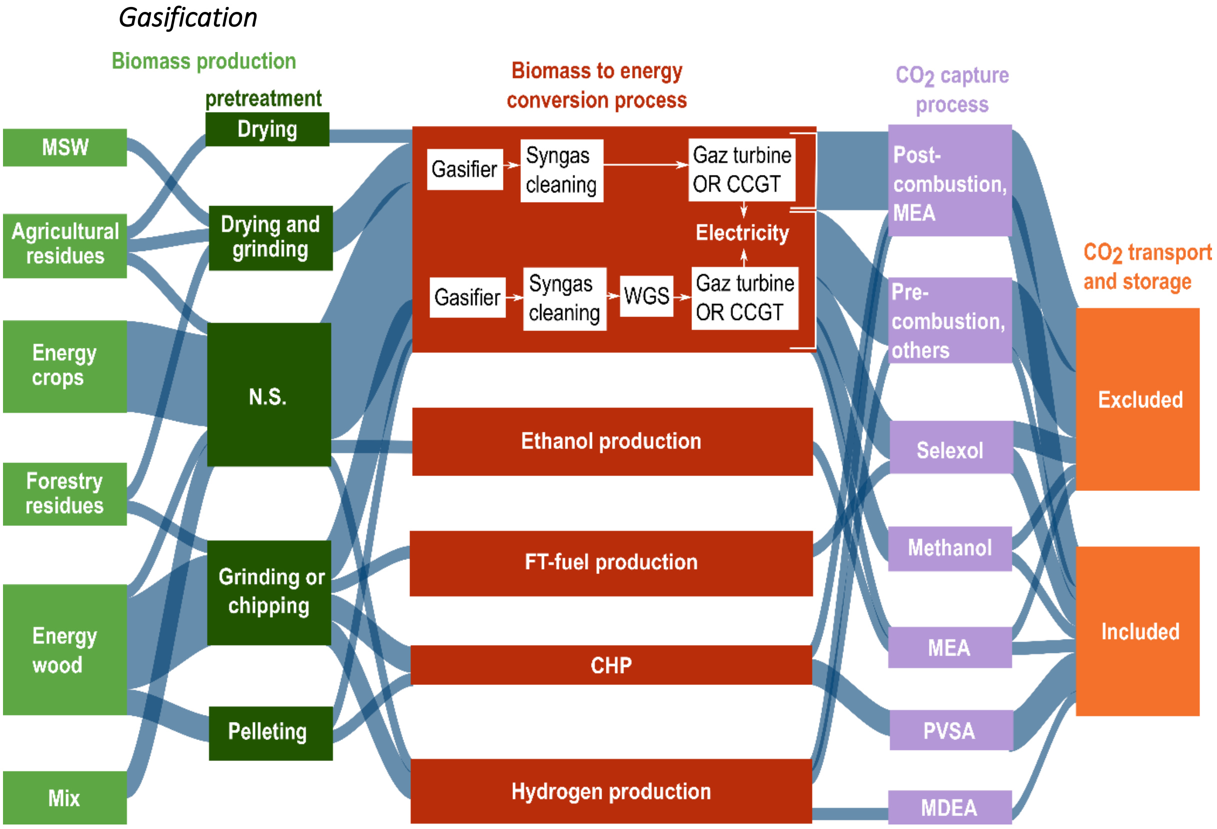

Figure 7: Overview of the systems studied in the 35 LCA of BECCS that include gasification of biomass. The thickness of a flow between two options is proportional to the amount of case studies that include the two options. CHP: Combined Heat and Power; CCGT: Combined Cycle Gaz Turbine; FT: Fischer-Tropsch; MDEA: Methyl diethanolamine; MEA: Monoethanolamine, MSW: Municipal Solid Waste; n.s.: not specified; PVSA: Pressure Vaccum Swing Adsorption; WGS: Water Gas Shift

As shown in Figure 7, the main studied function of the gasification process is the production of electricity. Processes involved to produce electricity through gasification of biomass are presented in Figure 7 depending on the chosen capture system. The values provided in Table 7 concern gasification without CCS as a mean of electricity production. They do not represent a specific type of gasifier. No consumption of heat is included because heat integration is performed. In fact, the syngas temperature is an important factor for optimising the efficiency of the gasification processes. For instance, it often needs to be cooled down prior to CO_2 capture. The heat in this case is not lost but recovered during the process (Bressanin et al. 2021; Liu et al. 2017; Zang et al. 2020). The excess heat produced can even be exported (Jana and De 2015; Jana and De 2016). As shown on Figure 7, pre-combustion capture occurs only in systems which include a water-Gas-Shift (WGS) step. During the WGS reaction, CO and H_2O are transformed into CO_2 and H_2. This reaction has two benefits: raising the concentration of CO_2 and thus the efficiency of the capture process (Cumicheo et al. 2019), and raising the LHV of the syngas. However, it consumes steam and catalysts (Antonini et al. 2020). If the goal is not to burn the syngas to produce heat and/or electricity, the composition of the syngas produced can be found in Larsson et al. (2021). More detail about the process is available in the SM.

In Table 7, the chosen bed materials are olivine and MgO because they are the only quantitative data available. However, bed materials could be silica sand (Jana and De 2015) or limestone (Antonini et al. 2021). Catalysts are also needed but data is limited. Antonini et al. (2020) include a list of catalysts in their inventory. However, the use of each catalyst is not clearly specified. Bressanin et al. (2021) indicate a consumption of 40.5 mg NiMgK/kg feedstock for tar reforming. Antonini et al. (2020) indicate a consumption of 18.97 mg NiO/kg feedstock and 2.61 mg MgO/kg feedstock. In the work of Antonini et al. (2020), ZnO is used both for desulfurization and low-temperature WGS (total: 34.73 mg/kg feedstock). In the work of Bressanin et al. (2021), desulfurization is carried out using H_2SO_4, water and DEA. For Antonini et al. (2020), the other catalysts are for low-, and high-, temperature WGS: CuO (33.88 mg/kg feedstock) + ZnO, and Fe_2O_3 (29.21 mg/kg feedstock) + Cr_2O_3 (3.37 mg/kg feedstock) respectively. Concerning the modelling of infrastructures, data is available in Zang et al. (2020). In Cavalett et al. (2018) and Oreggioni et al. (2017), infrastructures are modelled thanks to the Ecoinvent dataset (v2.2) “Cogen unit 160kWe, components for electricity only/ RER/ unit”.

Table 7: Gasification of biomass for electricity production - synthesis inventory table. kg feedstock as received in the power plant

	Type of value	Nb obs.	Year	Value	Min-Max	Unit
Parameters						
Electricity efficiency	mean	6	2020-2021	39.41	35-41	%
Capacity factor	mean	4	2004-2021	78.73	70-90	%
Plant lifetime	mean	9	2004-2021	26[a]	20-30	yr
Inputs						
Biomass				1.00		kg
Water						
- cooling	simulation	1	2021	0.04	-	kg/kg feedstock
- steam - gasification	mean	5	2011-2021	0.41	0.2-3.5	kg/kg feedstock
- steam - WGS	mean	2	2017-2021	0.16	0.15-0.17	kg/kg feedstock
- losses (steam cycle)	simulation	1	2015	0.11		kg/kg feedstock
Bed material						
Olivine	literature	1	2011	19.23		g/kg feedstock
MgO	literature	1	2011	3.41		g/kg feedstock

	Type of value	Nb obs.	Year	Value	Min-Max	Unit
Catalysts	see section 3.2.2.2					
Infrastructures	see Zang et al. (2020)					
Outputs						
Electricity	=input biomass*LHV biomass*efficiency					
Emissions to air						
CO_2	=C content of the biomass*44/12*input of biomass					
CO	simulation	1	<2015	121		mg/kg feedstock
CH_4	literature	1	<2011	122		mg/kg feedstock
N_2O	literature	1	<2011	80		mg/kg feedstock
NO_x	[b]	2	2017-2020		155-1880	mg/kg feedstock
H_2S	calculation	1	2017	16		mg/kg feedstock
SOx	[b]	2	2017-2020		5-720	mg/kg feedstock
PM	mean	2	2017-2020	37	18.6-43.4	mg/kg feedstock
Waste						
Ash	=fraction of ash*input biomass					
Sulfur	depends on biomass sulfur content					
Tar	mean	1	2017	61	50-71	mg/kg feedstock

[a] only three values: 20, 25 and 30 years

[b] large value range and only two observations.

Fermentation

Alcoholic fermentation directly produces high purity CO_2 (96% (Yang et al. 2021; Yang et al. 2020)). Previous studies therefore assumed that CO_2 capture is easier and only requires CO_2 compression. Only Bello et al. (2020) provided a complete LCI of the ethanol production according to Kautto et al. (2013)(see SM). Furthermore, the yields for ethanol production based on maize stover, miscanthus, switchgrass, poplar and hardwood are given in four different publications, ranging from 20 wt% to 24 wt% (dry basis). Inventories for ethanol production from corn and corn stover are available in the GREET® database (Wang et al. 2021). The inventories for ethanol production from wheat grain, maize, barley, rye, triticale, sugar beet, sugar can, straw are provided by the JRC (Edwards et al. 2019).

3.3.3 CO_2 capture

Table 8 presents an overview of the provided information and missing data on CO_2 capture processes. The LCI data for biomass to energy conversion process are given for the case where CCS is not implemented. The combination of a conversion process with a CCS process has two consequences:

1. A reduction of pollutants other than CO_2 in the flue gas. The reduction factor indicated can then be simply applied. In the particular case of oxy-fuel combustion, inputs linked to NOx treatment need to be deleted because NOx are not produced.
2. Loss in the efficiency of the plant if it is used for providing energy for the capture process. The efficiency with CCS (η_{CCS}) is equal to $\eta_{CCS} = \eta \times \left(1 - Q_{CO_2} \times E_{C02}\right)$, with η the efficiency without CCS, Q_{CO_2} the quantity of CO_2 produced by unit of energy and E_{C02} the energy to capture 1 kg of CO_2.

Table 8: For all the capture processes studied in the selected LCA, quantitative overview of a few selected LCI data (efficiency, energy) and overview of the data availability for the rest of the LCI. TRL: Technology Readiness Level; MEA: monoethanolamine, DEA: diethanolamine, MDEA: methyldiethanolamine, PVSA: Pressure Vacuum Swing Adsorption

Capture process	Amine-based absorption			Pre-combustion capture						Oxy-fuel
	MEA	DEA + LO-CAT	MDEA	Selexol	Rectisol	PVSA	Hot potassium carbonate	Calcium looping	Membrane	
TRL[a]	9	9	9	9	9	9	9	6	7-8	7
Number of case study	16	1	1	1	2	4	1	1	2	2
CO_2 purity, in	~16%	n.s.	n.s.	n.s.		~14%	n.s.	n.s.	n.s.	95%
CO_2 purity, out	>99%	n.s.	n.s.	n.s.	98%	90-99.9%	n.s.	n.s.	87%	
Efficiency	~90%	92%	98%	95%	94-100%	90%	95%	n.s.	90%	90-95%
Energy consumption										
Electricity, GJ/tCO_2	0.38	0.0	n.s.	0.1	0.1	1.1	n.s.	n.s.	n.s.	n.s.
Heat, GJ/tCO_2	3.69	3.6	n.s.	0.0	0.1	0.0	n.s.	n.s.	n.s.	n.s.
Chill, GJ/tonCO_2		?	n.s.	1.0	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.
Other inputs (-: not concerned, 0: data not available, 1: data in article)										
Infrastructure	1	0	0	0	0	1	0	0	0	0
Reagent makeup	1	1	1	1 (negligeable)	0 (methanol)	0 (sorbent)	0	0 (bed material)	0 (membrane)	0 (O_2)
Co-products/waste	1	1 (sulfur)	-	-	0 (sulfur)	-	-	-	0 (membrane)	-
Other emissions reductions	1	1	-	-	-	1	-	-	-	1 (NO_x)
Other inputs	1	-	-	-	-	1 (water)	?	-	-	-

a. TRL 9: "actual system is proven in an operational environment", TRL 8: "System complete and qualified", TRL 7: "System prototype demonstration in an operational environment", TRL 6: "Technology demonstrated in a relevant environment" (Bui et al. 2018; Bhadola 2020)

The most studied process for CO_2 capture is MEA-based capture, cf. Figure 4. MEA-based capture process is described in detail in the SM. The high energy requirement indicated in Table 9 is due to solvent regeneration. The building of the infrastructure has a low impact for all impact categories, apart from ecotoxicity (Koornneef et al. 2008). Koornneef et al. (2008) provide a LCI for infrastructure building. No default values are proposed for NOx and PM removal efficiency, because the range of values is too large, with a very low number of samples (less than 3 values).

Table 9: MEA-based CO_2 capture - synthesis inventory table. tCO_2 = tCO_2 captured

	Type of value	Nb obs.	Year	Value	Min-Max	Unit
Parameters						
Capture efficiency	mean	>14	2004-2018	89.80	75-100	%
Sorbent concentration	[a]	8	2013-2020	30.00	30-40	wt%
CO_2 purity						
- raw gas	mean	2		16.00	15-17	%
- final product	mean	6		99.00	99-99,8	%
Discharge pressure	mean	2		12.40	5.86-15.16	Mpa
Removal efficiency						
- Nox		3	2002-2019		1,25-90	%
- SO_2	mean		2002-2019	98.75	98-100	%
- PM		2	2002-2019		50-98	%
Inputs						
Solvent reclaimer - purification						
NaOH		1	<2006	0.13		kgNaOH/tCO_2
Activated carbon		1	<2006	0.075		kgC/tCO_2
Solvent losses						
- solvent	mean	12	2002-2021	2.05	0,27-3,98	kgMEA/tCO_2
- water	simulation	1	2020	0.63		tH2O/tCO_2
Energy requirement						
Heat	mean	11	2002-2017	3.69	2.95-7.52	GJ/tCO_2
Chill	mean	>3	2006-2021	103.50	91-106	tH20/tCO_2

	Type of value	Nb obs.	Year	Value	Min-Max	Unit
Electricity						
- pumping	mean	4	2005-2020	0.03	0.017-0.15	MWh/tCO_2
- compression	mean	4	2014-2020	0.07	0.02-0.11	MWh/tCO_2
Infrastructure	see Koornneef et al. (2008)					
Outputs						
Captured CO_2	= CO_2 released by energy production*efficiency of the capture					
Ammonia, to air	formula	5	2005-2021	286.32	38-555	g/tCO_2
MEA, to air	mean	12	2008-2020	40.78	0-117	g/tCO_2
CO_2, to air	= CO_2 released by energy production*(1-efficiency of the process)					

[a] the indicated value is not a mean as all but one article consider a MEA concentration of 30 wt%.

3.3.4 CO_2 Transport and Storage

CO_2 transport by pipeline and storage in an underground reservoir is often modelled by the consumption of electricity for CO_2 compression (transport and well injection) and possible leakage of CO_2, as indicated in Table 10. The construction of infrastructure (pipeline, well) is neglected for two reasons: i) the reuse of existing facilities, and ii) its negligible impact on climate change. An LCI of pipeline construction is available in the SM of Antonini et al. (2020).

The formula applied to calculate the energy requirement for transport was proposed by Damen et al. (2006) who worked on hydrogen compression and transport (see SM). The provided value is the electricity requirement to compress from 1 to 110 bar using a 4-step compressor. Depending on the capture process used and on the resulting CO_2 stream pressure, this electricity requirement could be reduced. Bello et al. (2020) assumed that no recompression is needed if the CO_2 travels less than 200 km in a pipeline. If the CO_2 is liquefied for ship-based transport, a supplementary energy consumption would have to be added (Bui et al. 2018).

Table 10: CO_2 transport and storage – synthesis inventory table

Input/Output	Type of value	Nb obs.	Year	Value	Min-Max	Unit
Transport						
Electricity	formula	7	2008-2020	111	21-111	kWh/tCO_2
CO_2 leaks						
- compressor		1	2006	23.24		tCO_2/MW/an
- pipeline		1	2006	2.32		tCO_2/km/an

Input/Output	Type of value	Nb obs.	Year	Value	Min-Max	Unit
Storage						
Electricity	formula		2008	7.00		kWh/tCO_2
CO_2 leaks	mean	3	2008-2020	0.07	0.03-0.1	%

4 DISCUSSION

An overview of the LCI data used in the selected LCA of BECCS is presented in the results section, through i) a review of the reproducibility, the completeness and sources of the data used across the selected articles, ii) when possible, synthesis inventory tables containing data years of production and data variability ranges. A lack of transparency concerning data quality and a lack of data availability have been observed. These issues were also highlighted by Terlouw et al. (2021) and Jeswani et al. (2022). There are methods to assess data quality, such as the pedigree matrix (Edelen and Ingwersen 2018). However, it has not been applied in the studied articles. Only a third of the articles (13/35) provide life cycle inventory tables. Half of the articles only assess the impact on climate change leading to inventory cut-offs. The representativeness, completeness and reproducibility of LCI data used in the selected LCA, as well as the relevance of the case studies for short-term deployments, are further discussed in the following sections.

4.1 LCI DATA QUALITY

4.1.1 Representativeness (temporal factor, data source)

Values for biomass harvesting and pretreatment essentially date from before 2016. In the pedigree matrix (Edelen and Ingwersen 2018), the use of data that are more than 6 years old corresponds to a quality grade for the temporal consideration criteria greater than 3/5 (1/5 stands for the best quality). Therefore, to better evaluate 2022 systems, the energy requirement values for harvesting (before 2016) and the required electricity for grinding (before 2013) would need to be updated. This also concerns: i) bed material consumption and infrastructure for gasification (before 2011), and ii) consumption of activated carbon and NaOH in MEA-based capture (before 2006). The foreground data collected in this review is relevant to model current and not future technologies. None of the studied LCA is described as a prospective LCA. Prospective LCA is still a field of LCA under development. Several frameworks or guidelines have been published in recent years (e.g. (Thonemann et al. 2020; Arvidsson et al. 2018; Cucurachi et al. 2022). Prospective LCA regroups LCA that aims to model future evolution of systems (Beloin-Saint-Pierre et al. 2020). Fajardy et al. (2017), Kar et al. (2021), Hammar and Levihn (2020) and Sproul et al. (2020) built temporally distributed LCI, in order to perform dynamic impact assessment on climate change. These temporally distributed LCIs aims to represent either the life of a power plant from its construction in the present to its future decommissioning or the dynamic of biomass growth. Thus, these temporally distributed LCIs do not consider technological evolution of foreground processes. It can be relevant to consider technological evolution notably for low TRL technologies, such as second generation capture process. If the goal is to evaluate a future deployment of actual low TRL technologies, LCI data can be adapted using scale-up procedures (Thomassen et al. 2019; Tsoy et al. 2020).

In every article, at least one step of the BECCS system is either not modelled or modelled from previous literature or background databases (quality grade of 4 or 5 in the updated pedigree matrix proposed by Edelen and Ingwersen (2018)). When process simulation is used to model the biomass to energy conversion process and the CO_2 capture, the biomass production is then modelled using bibliographic data. And conversely, if a large data collection effort is made to model biomass production, the modelling of the rest of the BECCS system is less documented. Collecting high quality data on the whole value chain of a BECCS system can be very time consuming and may not be relevant relatively to the goal of the LCA. It is important to inform the reader of the impact of data quality on the results of the study and the possible limitations that arise from it. Data used for modelling combustion, gasification and MEA-based CO_2 capture are mostly representative of current technologies. The values of emissions provided in the studied articles for biomass combustion are consistent with the lower boundary provided by the Best Available Techniques (BAT) established by the European Parliament and Council (2021). However, care should be taken in selecting the plant capacity factor. The capacity factor is used for shifting from the total plant life impact to the impact per unit of product. It can be very different between a plant producing electricity on site for direct industrial consumption (90%) and a plant producing electricity for the grid. The capacity factor is the ratio of the true total amount of energy produced in a year to the maximum of energy that is produced by the plant in one year. It also corresponds to the ratio of operated hours to total hours in a year. The capacity factor for grid electricity production can be found on government websites. For instance, the annual capacity factor of wood power plant is about 60% in the U.S.A. (EIA 2022), hence the 62.3 to 91.3% range of values given in the synthesis Table 7. The heat requirement for MEA regeneration is consistent with measured values at pilot plant scale, between 3.5 and 4.2 GJ/tCO_2 (Vega et al. 2020). The range for MEA makeup is also consistent with field measurements, i.e. 1.6 kg/tCO_2 captured (Morken et al. 2017). However, a lack of robustness is observed for the values of two flows of MEA-based capture process. The flows of sodium hydroxide and activated carbon come from only one source, i.e. Rao and Rubin (2006). The results on data source (section 3.2.3) also reveal the lack of robustness of the data used to model the CO_2 transport and storage stage. The data is mainly based on two sources from 2008 and before.

Moreover, the value for CO_2 leaks in pipeline transportation is based on observed methane leaks. However, in the case of a CO_2 leak, the interaction of CO_2 with its environment can worsen the degradation of the pipeline structure more quickly than in the case of a methane leak, since hydrated CO_2 is more corrosive than methane (Vitali et al. 2022).Therefore the amount of leaks during CO_2 transport could be underestimated. No leakage was observed during the testing and monitoring of large scale CO_2 storage (not for Enhance oil Recovery purpose) during the past 20 years (Gal et al. 2019). However, leakage might occur in later years, for instance due to corrosion of closed oil or gas wells (IPCC 2005; Gholami et al. 2021). This issue can be solved by proper storage management (Gholami et al. 2021). There is thus still a lack of hindsight on long term storage (100 years and more).

4.1.2 Completeness

Inventories conducted to assess only the impact on climate change are often not appropriate for multi-criteria assessments. In fact, some publications directly provide elementary flows of CO_2 associated with the consumption of inputs, without providing the quantity of input consumed. Consequently, they do not include other emissions than GHG emissions and the lack of detailed inputs makes it impossible to calculate them. For example, biomass production emits pesticides and fertilisers to the soil, air or water. Pesticides have an impact on ecotoxicity and fertilisers have an impact on eutrophication and acidification. Another example is the degradation of MEA into NH_3 during CO_2 capture. NH_3 is then emitted into the air, with an impact on acidification (UNEP-SETAC 2019). Therefore, environmental issues need to be addressed as a whole to prevent environmental impact transfers from climate change to other impact categories. All material consumption should be included to perform a complete LCA of BECCS and assess a trade-off between impact categories. The consumption of materials (infrastructure, membrane, catalyst, solvent...) is often neglected. Even if Antonini et al. (2020) observed that the impact of infrastructure on climate change is negligible, Zang et al. (2020) and Antonini et al. (2020) also demonstrated that the building of infrastructures contribute to other impact categories such as acidification, human toxicity or resource consumption. This may also explain why the capture of CO_2 from alcoholic fermentation processes is not assumed to require further capture equipment. However, in Gubler et al. (2020) the authors indicate that CO_2 from fermentation is "usually washed with water and then passed through activated carbon purifiers". The inventory for carbon capture from fermentation might therefore be incomplete for a multi-criteria assessment. In addition, neglecting infrastructure because it is being reused can conceal the environmental impacts of necessary well revamping or pipeline repairs.

4.1.3 Reproducibility

Although inventory tables are a suitable format for transmitting the relevant data and for facilitating the reproducibility of the study, they are rarely used. Tables are a useful format for LCA practitioners:

i) to check the completeness of the inventory, ii) to keep track of data information (variability, data source, geography…), or even iii) to apply a methodology to calculate a quality score. To prevent bias, the unit should be clearly and unambiguously stated. For instance, precisions should be given i) if a % is for mass, volume, mol, LHV, HHV, ii) if the tCO_2 refers to CO_2 captured or treated CO_2 or even iii) if the ton of biomass has a dry or wet basis. Data that enable unit conversions should also be provided, e.g. biomass moisture to convert t_{DM} to t_{WM}. To favour the reuse of the inventory, allocation methodology and factors should also be clearly stated.

4.2 BECCS SYSTEMS: RELEVANCE AND MISSING CASE STUDIES

4.2.1 Relevance of studied BECCS systems for a short-term deployment

The studied BECCS systems are relevant for short-term deployments. There is no competing use between implementing carbon capture for CO_2 storage and current CO_2 use. The Global CCS Institute (2022) identified only two commercial BECCS that are currently in operation and about 40 commercial BECCS projects. These projects aim to be operational by 2025 or earlier. Most of them are CCS applied to American ethanol plants. Currently, in the U.S.A., only approximately 25% of the CO_2 produced by ethanol production from fermentation is captured (Gubler et al. 2020). In Western Europe, according to internal data from the BIOC4M project (Lorne 2022) and the Gubler et al. (2020) market study only 10% of ethanol plants are equipped with carbon capture for CO_2 usage. Moreover, presently in the field of anaerobic digestion, CO_2 from biogas purification is generally released to the air (Gubler et al. 2020).

The types of biomasses studied in the selected articles all belong to the categories of biomass that have been evaluated as available for bioenergy by 2030 (Brown et al. 2020): agricultural residues, forestry residues and energy crops. However, algae cultivation is still facing scaling-up challenges (e.g., competing species contamination, oxygen inhibition, costly separation, and purification steps), which may explain why studies which chose algae as a feedstock are so scarce.

Combustion and gasification are the most studied biomass to energy conversion processes, possibly because they have already been deployed in certain countries (e.g. UK (García-Freites et al. 2021; Mohamed et al. 2021; Fajardy and Mac Dowell 2017)). Combustion of biomass is a cost-efficient and mature technology (Creutzig et al. 2015) for producing heat and electricity. TRL of gasification varies between 5 (Brown et al. 2020) and 9 (Shahbaz et al. 2021), depending on process configuration. Thus, gasification producing either syngas, or heat and electricity is ready for commercialisation (Shahbaz et al. 2021). Gasification followed by Fischer-Tropsch synthesis to produce advanced biofuels is soon to be industrialised (Brown et al. 2020), such as the BioTfueL® technology (IFP Energies Nouvelles 2021).

And syngas fermentation is still at a prototype stage (Brown et al. 2020).Gasification is not hindered by variations in the composition of the feedstock. As it is often the case with biomass, this is a valuable advantage.

MEA-based capture is currently a commercially available solution, and it is thus relevant for near-future deployments of BECCS. According to the 2020 report of the European Joint Research Center on CCUS (Kapetaki and Schleker 2020), post-combustion amines on power plant is the only CO_2 capture process that reached TRL 9 with pre-combustion NG processing. It has a high efficiency and selectivity for a low cost (IPCC 2005). The main advantage of such post-combustion processes is that they can be retrofitted in existing plants (Godin et al. 2021). However, research is still ongoing, particularly in the improvement of energy requirements and solvent losses that hinder their deployment (Kapetaki and Schleker 2020; Raynal and Tebianian 2020).

The two practical solutions for transport of large quantities of CO_2 over potentially long distances are pipelines and ship-based transport, or a combination of both (Bui et al. 2018). The two main options for geological CO_2 storage are saline aquifers and depleted reservoirs (Ineris 2017). Estimates of storage capacities are given in the IPCC Special Report on CCS (2005). However, it is stated that these estimates do not take into account the economic and technical feasibility of storage, as well as its sustainability (environmental and social). They should therefore be treated with caution. Thus, saline aquifers offer in theory large storage capacities between 1000 $GtCO_2$ (lower estimate) and possibly 10^4 $GtCO_2$ (IPCC 2005). However, injection of CO_2 leads to risk of overpressure within the geological structure. This risk is lessened in depleted reservoirs where CO_2 replaces the oil or gas that were present before extraction (Ineris 2017). However, the pressure exerted during oil or gas extraction can create fractures thus weakening the containment capacity of the storage. The allowable pressure for CO_2 storage without leaks is then reduced compared to the historical pressure of oil and gas storage. In addition, the presence of old wells represents another risk of CO_2 leakage if they have not been properly plugged. Both risks can be avoided by proper well and reservoir management. The storage capacity of depleted reservoirs is estimated between 675 $GtCO_2$ and 900 $GtCO_2$ (IPCC 2005).

4.2.2 Missing case studies

Further studies on CCS applied to stand-alone anaerobic fermentation should be performed to assess its potential for negative emissions. Indeed, anaerobic fermentation produces biogas with a high content of CO_2, which is thus more easily captured. The produced biogas essentially contains CH_4 (45 to 70 vol%) and CO_2 (30 to 55 vol%) (Escudie et Cresson 2017). Moreover, biogas upgrading is considered to be a source of CO_2 in nine out of the 56 LCAs on carbon capture and utilisation reviewed by Thonemann (2020b).

Second-generation capture processes are still under development and also need to be better assessed as part of a BECCS system. For instance, they include phase change solvents, chemical looping combustion or membrane, while first generation processes include absorption (amine-based solvents), Temperature or Vacuum Pressure Swing Adsorption (TSA or VPSA, solid adsorbent) processes and oxy-fuel capture (Raynal and Tebianian 2020). The second-generation capture processes are developed to deal with the high energy requirements of first generation processes.

A wider range of options for transport and storage of CO_2 should be assessed and compared. Indeed, since BECCS systems are limited by the availability of biomass, the amount of CO_2 to be transported may be low enough for competitiveness between rail and pipelines. Other storage possibilities also exist, such as coal steams (adsorption), basalts (mineral trapping), salt caverns (physical trapping), abandoned mines and mineral carbonation (IPCC 2005). Using these storage options could put CO_2 storage in competition with other uses such as coal mining for coal steams (IPCC 2005) or hydrogen storage for salt caverns. The storage capacity of these solutions has yet to be established. Despite all, these niche options could provide solutions for geological storage of CO_2 in some projects and may then deserve a thorough assessment.

4.3 LCI SYNTHESIS TABLES: USE AND LIMITATIONS

LCI synthesis tables were build based on the collected LCI data from the 35 LCA on BECCS (see section 3.3). The application scope of the LCI synthesis tables is specified in the following first subsection. Their limitations are specified in the second subsection.

4.3.1 Application scope

Collecting LCI data is a time-consuming step in LCA. Due to time and data availability constraints and the LCA goal, not all steps of a BECCS system may be modelled with the same accuracy. For instance, Lausselet et al. (2017) studied existing Norwegian Waste-to-Energy plants, with the goal of comparing four scenarios including a scenario where CCS is added to the power plants. The main modelling effort was put on collecting data on the 17 existing plant and model their direct emissions based on the waste composition. Lausselet et al. (2017) then modelled the CCS step using data from the literature (Koornneef et al. 2008; Rao and Rubin 2002). In this case, generic LCI tables can be helpful. However, such synthesis tables are not suitable to model a foreground process with high contribution to the total impact of the system. LCA is an iterative methodology. A LCA practitioner can use a synthesis table in the initial modelling to fill a data gap. If the results then reveal a high contribution to the total impact of the system of the process modelled with a synthesis table, the LCI needs to be improved, or the results must be used with caution.

The synthesis LCI tables can also be used for LCI analysis. The synthesis LCI tables provide a state-of-the-art of the values used to model BECCS system, including variability range across studies. Therefore, the synthesis LCI tables can be used to compare new measured or calculated LCI data to the existing literature. Moreover, the provided variability ranges can be used to include uncertainty in the LCA, by performing either sensibility analysis on chosen inputs or a Monte Carlo analysis.

4.3.2 Limitations

The provided LCI tables are meant to be a state-of-the-art of the data used in LCA of BECCS, which is obviously a limited literature coverage. It explains why the LCI tables are potentially based on few data points (mostly below 5). The LCI tables could be improved by using data from a wider corpus of articles, e.g. LCA on bioenergy without CCS or literature on process simulation, but this is outside the scope of the study.

LCA practitioners should be aware that correlations exist between the inputs and outputs of each process described by the synthesis LCI tables. Additional knowledge to better represent the technical specificity of the processes should be used when available to the practitioner. For instance, empirical estimations exist to correlate grinding electricity consumption with particle sizes as shown in Onarheim et al. (2015) for grinding of wood. Another example is the correlation between the heat demand for MEA-capture and several process parameters, such as CO_2 capture efficiency or sorbent concentration (Rao and Rubin 2006). Therefore, LCA practitioners should prefer measurement or process simulation to the provided synthesis LCI tables when possible.

5 CONCLUSION

The goal of this study is to review, from a quantitative and qualitative point of view, the LCI data used in LCAs of BECCS. The LCI data of 35 LCA of BECCS have thus been inventoried. Results highlight insufficient discussion concerning the relevance of LCI data quality relatively to the LCA goals in the studied articles. Studies on variability and uncertainties are carried out, but other quality criteria data such as geography, temporal factors or consistency were generally omitted. A scarcity in data availability was also observed. Providing LCI data is essential to ensure the reproductivity of an LCA. It can be hindered by industrial secrecy when data are based on a given plant. However, this review reveals that literature data or simulation data are the main data sources. LCI data should thus be more available. For almost half of the studies, only climate change has been assessed. This leads to incomplete LCI data for performing a multicriteria analysis. Indeed, consumption of materials, building of infrastructures or emissions other than GHG were neglected. However, for example, as the issue of resource scarcity becomes increasingly significant, catalysts should also be added to the assessment.

The search query defined in section 2 was extended to May 2023. Since 2022, 14 LCAs on BECCS have been published (see SM). None of these articles contain a subsection dedicated to a discussion on the quality of the LCI according to the criteria of ISO 14044 and its consistency with the LCA goals. Half of the articles provide inventory tables, showing a slight improvement in data availability. However, nine articles still evaluate the impact only on climate change.

Based on the collected LCI data, synthesis LCI tables for most BECCS processes were built, including biomass harvesting, pretreatment, combustion, gasification, and MEA-based CO_2 capture. The inventory tables are site-generic and represent current technologies. The inventory on MEA-based CO_2 capture represents a step towards filling the gap in the Ecoinvent database concerning CO_2 capture processes. Moreover, all the LCI synthesis tables provide variability ranges that allow for relevant uncertainty analyses to be performed. These LCI tables can also facilitate harmonisation between future LCAs on BECCS, by providing data to maintain similar system boundaries and assumptions despite varying LCA goals. However, they should not replace measurement or process simulation when possible.

For further work on the subject, it would be noteworthy to search for LCA at each life cycle stage of a BECCS system (biomass production, bioenergy, CCS) and to complete the study with the obtained data. In the present review, the collected data are mainly representative of current technologies. However, certain data, such as energy requirements for harvesting or bed material consumption for gasification, deserve to be updated. Moreover, investigating scale-up procedure or other strategies to predict the evolution of BECCS technologies and its impact on LCI data would be of interest to evaluate the impact of future deployment (prospective LCA approach). Some BECCS case studies should also be evaluated in more detail using LCA. These case studies include processes such as stand-alone anaerobic fermentation, second-generation capture processes or other types of transport (i.e. rail) and storage options. This recommendation is maintained after browsing through the 14 newly published LCAs. None of these articles studied stand-alone anaerobic fermentation. Only one studied second-generation capture processes, i.e. post-combustion CO_2 capture using membrane. All the articles including CO_2 transport in the system boundaries considered pipeline for CO_2 transport. Lastly, an evaluation of the environmental benefits of using CO_2 before permanent storage would also be worthwhile.

CHAPTER CONCLUSION

This review revealed several issues concerning data management in LCA that should be considered for the case study used in the next chapters:

- It emphasises a lack of availability of the inventory data. As far as transparency and reproducibility are concerned, all LCI data used in this work are provided as supplementary materials of the articles in the form of excel files, together with the scripts used to calculate the LCA in Brightway2.
- The majority of the reviewed articles did not discuss the adequacy of their inventory data quality in relation to the purpose of their LCA. In this thesis, the case study illustrates the methodological issues raised in the chapters 2 and 3. The results are not intended to be used to decide whether the case study should be deployed. For this reason, the discussion of the quality of the LCI data is not required, but the choice of the system is explained in below.
- The evaluation of negative emission potential requires "from-cradle-to-grave" system boundaries. Thus, the chosen case study needs to include a CO_2 source, a CO_2 valorisation process, and the end-of-life of the CO_2-based product.

The selection of the case study was driven by the availability of its inventory data, by the maturity of technologies for deployment by 2050, and by their adequacy to illustrate the methodological issues. The five commercial BECCS systems identified by the Global CCS Institute (2024 update) are all ethanol plants (alcoholic fermentation). Thus, ethanol plants are the most promising CO_2 source. Moreover, ethanol plants can use a wide diversity of biomass as feedstock through more or less complex biomass pre-treatments. It means that it is possible to perform a sensitivity analysis on the biomass used as a feedstock. Therefore, an ethanol plant is selected as the CO_2 source for the case study. Biomasses are selected to represent the diversity of available biomasses and the associated methodological challenges. Thus, maize is chosen as a representative of conventional biomass as it is currently one of the most common feedstocks for ethanol production at world scale. Wood residues and miscanthus are then chosen as representatives of lignocellulosic feedstocks for second generation ethanol pathway.

Concerning CO_2 valorisation, methanol is a major building block in the chemical industry and can be produced from CO_2 as indicated in the section 3 of the introduction. Methanol can either be used as a fuel or it can be converted into a wide range of products usually derived from petrochemicals. The technology to convert methanol to olefins (olefins: ethylene and propylene) is mature and already used on an industrial scale in China 68% of propylene is used to produce polypropylene. And polypropylene accounts for 19.4% of European plastics demand, according to Plastics Europe (2020). The meta-analysis of Thonemann (2020) on LCA of CCU is the main source of the LCI data used to model the case study. A reusable polypropylene shopping bag is chosen as the product because it is an everyday product. In France, in 2018, 43.3% of plastic waste was treated by incineration with energy recovery, 32.5% by landfilling and 24.2% by recycling (Plastics Europe 2020). The present review led to the compilation of generic LCI tables. The LCI tables for the CO_2 capture by MEA-solvent, and the transport and CO_2 storage, are used to model the end-of-life of the reusable polypropylene shopping bag in the case study: incineration with CCS.

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Chapter 2: Life cycle assessment of carbon capture and utilisation as a negative emissions technology: recommendations and case study

The previous chapter was linked to a first methodological choice in LCA: the definition of system boundaries. Thanks to the data collected, it is now possible to easily model a system with boundaries "from-cradle-to-grave". But assessing negative emissions raises other issues as i) taking atmospheric CO_2 into account, ii) defining the functional unit and iii) managing multifunctionality. In this chapter, the compatibility of the existing guidelines for LCA of CCU with the recommendations made in the literature for NETs is examined with regards to these three points. This chapter was submitted to the International Journal of Life Cycle Assessment the 05/02/2024 and is currently being revised: Duval--Dachary S., Lorne D., Beauchet S., Salou T., Hélias A. Life cycle assessment of carbon capture and utilisation as a negative emission technology: recommendations and case study.

List of abbreviations

CCU	Carbon Capture and Utilisation
CCUNET	Carbon Capture and Utilisation as Negative Emissions Technology
DDGS	Distiller's Dried Grains with Solubles
GHG	GreenHouse Gases
IPCC	Intergovernmental Panel on Climate Change
JRC	Joint Research Center
LCA	Life Cycle Assessment
LUC	Land Use Change

1 INTRODUCTION

In its latest report on climate change, the Intergovernmental Panel on Climate Change (IPCC 2023) highlights the threat posed by climate change to mankind, biodiversity and ecosystems in general. The IPCC stressed that reaching net zero CO_2 emissions, and even net zero greenhouse gas (GHG) emissions, is a necessary condition for limiting human-caused global warming. Carbon Capture and Utilisation (CCU) contribute to the transition towards a net zero society (Kapetaki and Schleker 2020), along with sufficiency and energy efficiency. CCU involves the recycling of captured CO_2, either from an industrial process or directly from the atmosphere, for purposes of further usage. The uses of captured CO_2, with or without its transformation (chemical or biological) can vary (Kapetaki and Schleker 2020), including plastic production. Products created from CCU processes are substituted for products generally created from raw materials of fossil origin (reference products). The environmental benefits of this substitution, assessed by comparing the impact of the production of the reference product with the impact of the production of the CCU-based product, are called avoided emissions.

The evaluation of the environmental benefits of CCU requires an assessment over the entire life cycle of the system, using Life Cycle Assessment (LCA). LCA is the standardised method to perform the environmental assessment of products and services over their whole life cycle. Numerous LCA of CCU systems have previously been published, as reviewed by Thonemann (2020). In 2020, both the European Commission and the CO_2 global initiative (Michigan University) published guidelines to harmonise LCA on CCU systems and improve the comparability of results (Ramirez Ramirez et al. 2020; Zimmermann et al. 2020). The two guidelines provide recommendations specifically for CCU systems at each step of an LCA: system boundaries, definition of the functional unit, choice of reference systems and so on. In 2021, the Joint Research Centre (JRC) published guidelines for carrying out LCA on plastics produced from alternative feedstocks, including CO_2 (Nessi et al. 2021). In the three documents, “negative emissions” or “carbon negativity” were only briefly mentioned.

CCU as a Negative Emission Technology (CCUNET) is a concept that is only beginning to emerge. NETs are designed to intentionally remove atmospheric CO_2 (carbon dioxide captured from the atmosphere by biomass growth or other processes) (Minx et al. 2018). Desport and Selosse (2022) provided a review of CCU systems that have the potential to generate net negative emissions. They then explored the potential of each system from technical and economic perspectives. They evaluated the global potential of trapping atmospheric CO_2 into plastic at 1.1 – 2.3 $MtCO_2$ per year. Aracil et al. (2023) focus on long-term storage of CO_2 as plastic. Using LCA, they demonstrated that renewable-derived plastics from municipal solid waste were NET (capture during production and long-term storage of atmospheric CO_2 in landfills).

Permanent storage of atmospheric CO_2 does not necessarily lead to net negative emissions, due to the consumption of energy and materials throughout the whole life cycle. To achieve net negative emissions, the impact on climate change calculated with LCA of the GHG emissions due to the system should not offset the benefit of the GHG removal (Tanzer and Ramírez 2019). This leads to the research question addressed in this article: "are guidelines for performing LCA on CCU compatible with the environmental evaluation of CCU systems designed to generate negative emissions?"

No exhaustive guideline exists for LCA of NET systems. However, articles discussing methodological issues linked to LCA of NET systems have been published. Goglio et al. (2020), Brander et al. (2021), Jeswani et al. (2022), Terlouw et al. (2021) and Zakrisson et al. (2023) all highlight the significance of choosing a relevant functional unit. Jeswani et al. (2022) and Terlouw et al. (2021) showed that no consensus prevails on the method to deal with NET systems producing multiple products or services, i.e. multifunctionality. Goglio et al. (2020), Brander et al. (2021) and Jeswani et al. (2022) discussed the issue of the timing of CO_2 removal and emission. The present article focuses on these three most discussed methodological issues, within the context of CCUNET systems:

- *Accounting for atmospheric CO_2.* This issue first needs to be solved before the timing of removal and emission can be dealt with. The current consensus in LCA is to consider that emissions of atmospheric CO_2 have no effect on climate change, since they were initially captured from the atmosphere.
- *Choosing the conventional counterpart.* The choice of the functional unit is placed within the context of selecting the conventional counterpart. Indeed, the amount of avoided emissions generated by the system strongly depends on the chosen conventional counterpart. For instance, the amount of avoided emissions generated by burning a CO_2-based synthetic fuel in a vehicle ought to be different if compared with a fossil-based fuel or an electric vehicle. Here, the term "conventional" is not synonymous for "fossil-based". For example, the conventional counterpart could also be a bio-based product.
- *Dealing with the multifunctionality*.

The overall aim is to identify the existing recommendations, check their relevance when a CCUNET system is evaluated, and propose new recommendations if necessary. The relevance of methodological choices should be evaluated relative to the purpose of the study. CCUNET has two main goals for which the relevance of CCU guidelines need to be evaluated:

- *Goal 1: Comparing with a conventional counterpart*. Zimmerman et al. (2020) concluded from a short literature review that the comparison of the CCU system with "the same product or service derived from fossil carbon source" is one of the most commonly intended applications for LCA of CCU. This conclusion is supported by a more comprehensive review carried out by Thonemann (2020). Ramirez et al. (2020) broadened the objective by strongly recommending a comparison with “conventional or other new products and services, which is called the “Reference system””.

- *Goal 2: Assessing whether net negative emissions are generated*. This is obviously an essential requirement for a NET system.

In the first section, the scope is specified for each goal. In the second section, the recommendations are applied to a case study in order to illustrate their relevance or issues. In the third section, recommendations are examined according to the functional unit or to the modelling of atmospheric carbon. These discussions lead to the formulation of recommendations specific to the CCUNET system to be used in addition to existing guidelines for the CCU.

2 MATERIALS AND METHODS

The existing recommendations are summarised in Table 11. Since no exhaustive guidelines exist for LCA of NET systems, the recommendations provided in research or review articles by Goglio et al. (2020), Brander et al. (2021), Zakrisson et al. (2023), Jeswani et al. (2022) and Terlouw et al. (2021) have been grouped into a single column.

In the first and second subsections, the functional unit and system boundaries are explained for goal 1 and goal 2, respectively. The choice of the conventional counterpart is explored through goal 1. The relevance of methods for dealing with the multifunctionality of the CO_2 source is examined through goal 2. In the last subsection, a case study is presented. The case study will be used in order to assess the relevance of the recommendations.

Table 11: Summary of the existing recommendations on the three main methodological points for LCA of CCU identified as open to discussion relative to the assessment of NET. LCI: Life Cycle Inventory, LCIA: Life Cycle Impact Assessment, CCS: Carbon Capture and Storage

	Recommendations for LCA of CCU			Recommendations for LCA of NET in research or review articles
	Ramirez et al. (2020)	Zimmermann et al. (2020)	JRC (Nessi et al. 2021)	
Accounting for atmospheric CO_2	Full accounting, in LCI and LCIA	Not specified	Accounting in LCI but not in LCIA	No accounting observed by Jeswani et al. (2022)
Choosing the conventional counterpart – Functional unit definition	The reference system should provide the same functions as the system under study. The functional unit should be defined according to the requirement of the ISO 14040/44 standards.			Goglio et al. (2020) recommended that the carbon sequestration and other secondary functions such as land occupation or income generation should be included in the functional unit. Zakrisson et al. (2023) recommended a sensitivity analysis to be performed on the chosen functional unit.
	"mass of CO_2 utilized" could be used to compare CCU/CCS systems		No complementary recommendation	
Dealing with multifunctionality (CO_2 source)	System expansion. (Consequential LCA) CO_2 = undesired side product	Allocation by physical causality. CO_2 = product	Circular Footprint Formula. CO_2 = waste for recycling	If substitution is used, avoided emissions need to be separated from negative emissions (Jeswani et al. 2022; Terlouw et al. 2021). Mass allocation allows for the calculation of meaningful negative emissions (Terlouw et al. 2021).

2.1 SCOPE OF GOAL 1 - COMPARISON WITH A CONVENTIONAL COUNTERPART

Before defining the conventional counterpart and system boundaries, the intended application of the LCA on the CCUNET system must be clearly established. Comparisons with conventional counterparts observed by Zimmerman et al. (2020) and Thonemann (2020) and recommended by Ramirez et al. (2020) share the same intended application, i.e. to answer the following questions:

- Is the studied system environmentally beneficial if it is compared with a business as usual situation?
- If the answer is yes, is it environmentally better (1) to capture, use and then permanently store the CO_2 (CCUNET) or (2) to capture and permanently store the CO_2 (CCS)?

The reference system should be relevant for the intended application but should also share the same functions as the studied system. The studied system is multifunctional, and this multifunctionality is solved by system expansion. The functions are supplied by the CO_2 source and the CCU system. The CO_2 source produces one or more products, for instance, in the case of bioenergy production, heat and/or electricity and/or fuels. A CO_2 source is not built specifically for CCU, or CCUNET, therefore, the same CO_2 source is included in the reference system, with or without carbon capture. Then for simplicity, the part of the functional unit related to the CO_2 source can merge with its reference flow, i.e. the same quantities of products are maintained between the compared systems. CO_2 can be considered as a product or as a waste, as indicated in Table 11. In the context of climate change, the aim is to reduce the amount of emitted therefore produced CO_2, and not to encourage extra production of CO_2. Moreover, except for very specific processes such as ammonia production, industrial CO_2 emissions are not currently captured but are instead released because they have no economic value (Nessi et al. 2021). CO_2 is thus considered to be a waste that must be reduced. The CCU system can also produce one or more products. The functional unit related to the products of the CCU system (e.g. reusable polypropylene shopping bag) is the sum of the functions envisaged for the products (e.g. carrying groceries). The reference system could then be the product originating from the conventional production route (e.g. fossil-based polypropylene shopping bag) or a totally different product answering the same function (e.g. cotton-based shopping bag). However, in comparison with other products that answer the same functions, the environmental relevance of the product originating from the conventional production route is assumed to have been validated by other studies and any problematic burden-shifting ought to have been identified. In this case, it is relevant to simply compare the CCU production route with the conventional production route. To conclude, the functional unit related to a CCUNET system includes: i) the amount of products from the CO_2 source and ii) the amount of products from the CCU system.

To answer the first question, ‘Is the studied system environmentally beneficial compared with a business as usual situation?’, the reference system should be adapted to the world as it is today. This has two consequences that are illustrated in Figure 8. Firstly, there is no carbon capture. The CO_2 is released into the atmosphere, at the CO_2 source and at the end-of-life of the conventional counterpart of the CCU system. Secondly, for the CCU system, the conventional production route is the fossil-based route. According to the IEA, the chemical sector is not on track with the Net Zero Emissions by 2050 scenario. Half of the chemical sector’s energy input is still used as raw materials (Perez Sanchez 2023). This implies that fossil resources are still a widely used feedstock in the chemicals sector.

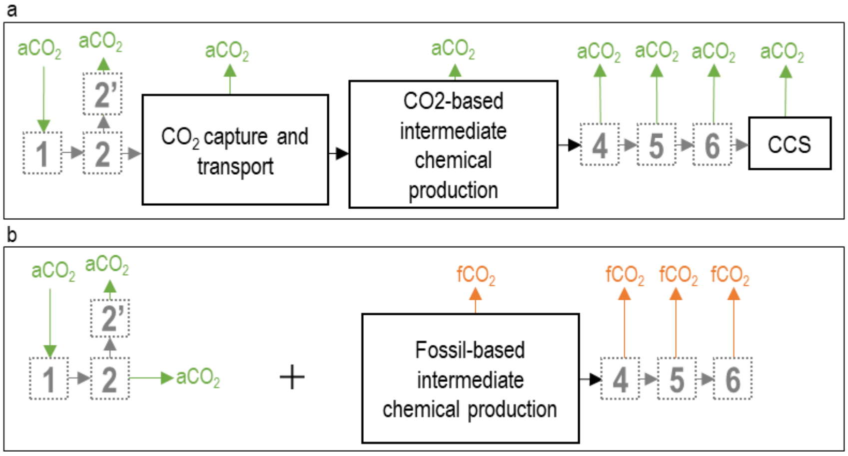

Figure 8: System boundaries in order to answer the question “Is the studied system environmentally beneficial when compared with a business as usual situation?”. Boxes with dotted lines correspond to steps that are identical for the CCUNET system (a) and for the reference system (b). aCO2: atmospheric CO_2, fCO2: fossil CO_2. 1: Biomass production, 2: CO_2 source, 2’: End-of-life of the products of the CO_2 source, 3: CO_2 capture, 4: Transformation of the intermediate chemical in a product, 5: Product use, 6: Product end-of-life without carbon capture and storage (CCS), 7: CCS

To answer the second question, ‘Is it environmentally better (1) to capture, valorise and finally permanently store the CO_2 (CCUNET) or (2) to capture and permanently store the CO_2?’, the carbon capture and storage (CCS) was added to both the CO_2 source and to the end-of-life of the reference system used to answer the first question, see Figure 9.

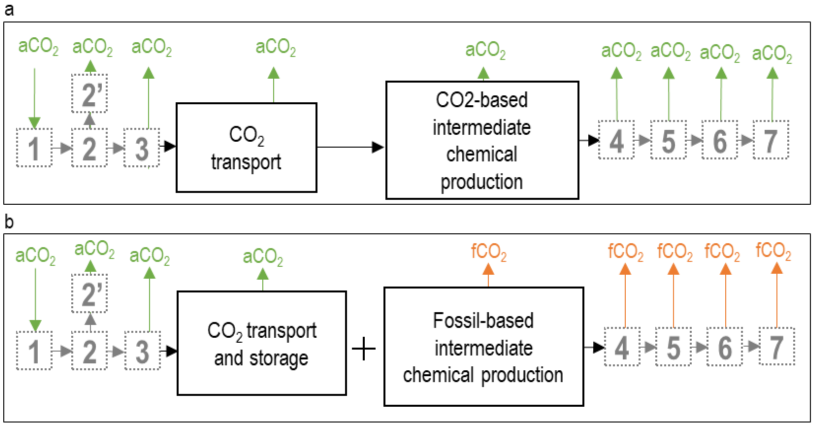

Figure 9: System boundaries in order to answer the question "Is it environmentally better (1) to capture, valorise and finally permanently store the CO_2 (CCUNET) or (2) to capture and permanently store the CO_2 (CCS)?". Boxes with dotted lines correspond to steps that are identical for the CCUNET system (a) and for the reference system (b). aCO2: atmospheric CO_2, fCO2: fossil CO_2. 1: Biomass production, 2: CO_2 source, 2': End-of-life of the products of the CO_2 source, 3: CO_2 capture, 4: Transformation of the intermediate chemical in a product, 5: Product use, 6: Product end-of-life without carbon capture and storage (CCS), 7: CCS

The system boundaries that were recommended for evaluating systems with negative emissions are "from-cradle-to-grave" boundaries (Tanzer and Ramírez 2019). As illustrated in Figure 8 and Figure 9, the CCUNET system shares identical unit processes with its reference system because of the two following assumptions:

- The CO_2 source was not specifically designed to perform CCU, it is identical between the two systems, and only the treatment of the CO_2 differs (captured or released).
- The CO_2-based product is assumed to have the same technical quality, including recyclability as its conventional counterpart.

These identical unit processes can be excluded from the system boundaries without distorting the comparison. Therefore, selecting a specific use and end-of-life for the intermediate product is not necessarily needed to answer the second question.

2.2 SCOPE OF GOAL 2 - ARE NET NEGATIVE EMISSIONS GENERATED?

The definition of a functional unit does not represent a challenge for this goal, since net negative emissions are an intrinsic property of the system. Net negative emissions, unlike avoided emissions, are not calculated in relation to a reference system. The recommended system boundaries for evaluating net negative emissions are "from-cradle-to-grave" boundaries (Tanzer and Ramírez 2019). All the life cycle steps must be included, from the capture of atmospheric CO_2 to either its reemission to the atmosphere or its permanent storage outside the atmosphere.

The system can produce multiple products, as demonstrated in the previous section. A more precise goal could be defined by the following question: “what amount of net negative emissions is generated by the CO_2-based product only?”. The answer to this question could then be re-employed as background data in another LCA. The answer would require multifunctionality to be solved, i.e. by identifying which fraction of the impact that could be allocated to each given product. The existing recommendations, summarised in Table 11, are recalled in Figure 10[1]. System expansion does not enable the more precise goal to be answered. It is nonetheless included in the method for solving multifunctionality. System expansion corresponds to an extension of the system boundaries until they encompass the complete life cycle of each co-product. System expansion allows for flows of atmospheric carbon to be monitored from CO_2 capture to release without distortion. System expansion has therefore been chosen as the reference method for assessing the net negative emissions potential.

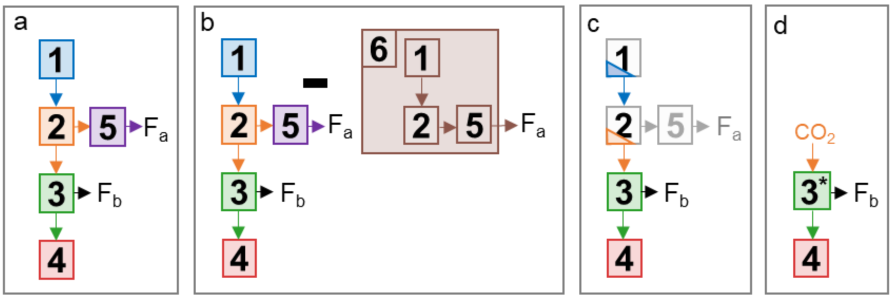

Figure 10: Options to deal with the multifunctionality of the CO_2 source. a: System expansion, b: Substitution or allocation by physical causality, c: Allocation (pale boxes = allocated to F_a), d: Circular footprint formula. F_a: Functions associated with the products of the CO_2 source, F_b: functions associated with the products of the CCU process. 1: Biomass production, 2: CO_2 source, 3: CO_2 to product, 3: CO_2 to product including the production of fossil-based intermediate product, 4: End-of-life of the CO_2-based product, 5: End-of-life of the CO_2 source products, 6: Avoided emissions*

2.3 CASE STUDY: INVENTORY AND IMPACT CHARACTERISATION METHODS

The case study is illustrated in Figure 11. It includes ethanol production as a CO_2 source and the production of CO_2-based polypropylene as a CCU system. It was considered to be a potential CCUNET system because the CO_2 initially captured by the biomass is permanently stored at the end-of-life of the reusable polypropylene shopping bag. This was chosen as a case study because the CO_2 source is multifunctional, the inventory data are available in the literature, and it is coherent with existing technologies. The inventory data were obtained entirely from the literature. The background datasets were from Ecoinvent v.3.8 (Moreno Ruiz et al. 2021) and Agribalyse v3 (ADEME). Whenever possible, France was chosen as the production location. If not, the locations were European or Global. Goal 1 was split into two questions (see section 2.1), which define the two reference systems, see Figure 8

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[1] See definition ‘Multifunctionality’ in section “Keys definitions, acronyms and abbreviations” for more details

and Figure 9. For both reference systems, the fossil-based intermediate chemical was fossil-based propylene modelled with an Ecoinvent dataset ('market for propylene [RER]') (Moreno Ruiz et al. 2021). The aim of this case study is to illustrate methodological issues and should not be used for deciding whether or not to deploy this specific CCUNET system. Certain assumptions were made to streamline the system. No impact was considered for the bag use phase. No CO_2 transport was considered between the CO_2 source and the CCU process. The infrastructure and process to capture CO_2 from ethanol production were not modelled and their impacts were assumed to be negligible (Duval-Dachary et al. 2023).

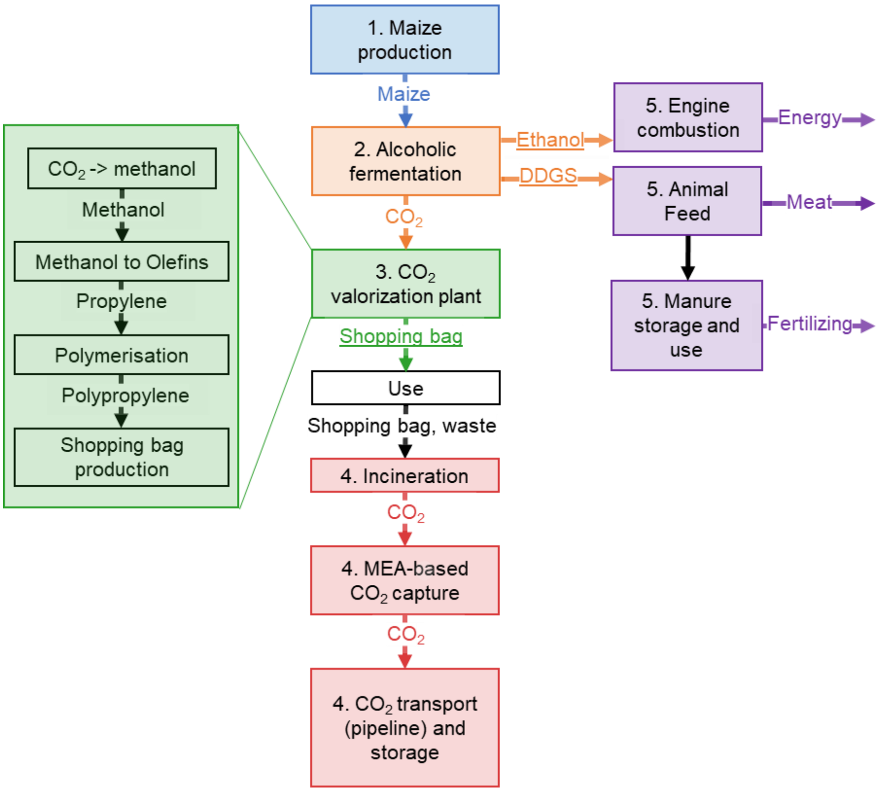

Figure 11: Life cycle steps included in the case study. Products underlined = main products. DDGS: Distiller's Dried Grains with Solubles. MEA: Monoethanolamine. 1: Biomass production, 2: CO_2 source, 3: CO_2 to product. 4: End-of-life of the CO_2-based product, 5: End-of-life of the CO_2 source products

To perform allocation, the carbon content was chosen as a physical factor, in order to maintain the mass balance of atmospheric CO_2. The allocation factor can thus be calculated as the ratio between the mass of carbon in the product and the total mass of carbon contained in all the products of the process. To apply the circular footprint formula, the point of substitution is defined as the moment when the process becomes identical between the CO_2-based product and the fossil-based product (Nessi et al. 2021). The point of substitution is reached when propylene is produced by methanol to olefins. Therefore, the production of primary material corresponds to the production of fossil-based propylene. The quality ratio is assumed to be 1, i.e. the CO_2-based propylene has the same quality as fossil-based propylene, in the absence of specific information (Nessi et al. 2022). The allocation factor between supplier and user is assumed to be 0.5 because there is currently no established market for CO_2-based propylene, and this is the default value for polypropylene proposed in appendix C of the Plastic LCA method (Nessi et al. 2021; Nessi et al. 2022). However, as pointed out by the JRC, "any future study focusing on real products shall take into account the specific market situation at the time of the study itself." CO_2 is currently considered as a waste, thus implying that none of the impacts arising from activities before CO_2 capture are attributed to the CO_2-based product.

A full evaluation of atmospheric CO_2 was selected ("+1/-1" approach), as recommended by Ramirez et al. (2020). Flows of captured CO_2 were included in the inventory with a negative value, i.e. negative flows, and a characterisation factor of 1, leading to a positive effect on climate change. This choice is discussed in section 4.3. Accounting for atmospheric CO_2 has an impact on inventory modelling and impact characterisation. Firstly, it raises challenges for inventory modelling, in particular for the biomass production step and for the end-of-life of Distiller's Dried Grains with Solubles (DDGS). In the biomass production step, a negative flow of atmospheric CO_2 was added according to the carbon content of maize ("Carbon dioxide, non-fossil"). A "Carbon dioxide, in air" flow had already been included in the Agribalyse dataset chosen to model maize production. However it was removed to avoid double counting. For the DDGS end-of-life step, DDGS was assumed to serve as animal feed (Buenavista et al. 2021). A carbon mass balance was performed to identify the amount of ingested carbon released to the atmosphere in the form of CO_2 and of CH_4. Due to limited existing publications on the subject, the calculation was performed for ruminant feed only (Lecomte et al. 2004). For 1 kg of ingested DDGS, around 0.7 kg is emitted as CO_2 and 0.3 kg as CH_4. This includes emissions due to ruminant breathing and digestion as well as the management of manure and its use as fertilizer. To avoid double counting, emissions resulting from the application of organic fertilizer during maize production were not counted. For further details on the calculation, see the online resource ESM1. Secondly, the impacts are characterized using EF 3.0 methods (Fazio et al. 2018), which do not fully include atmospheric carbon. The characterisation factors of the "climate change" and "climate change, biogenic" impact categories were thus modified to fit the "+1/-1" approach. A symmetry in the response of the climate system between CO_2 emissions and capture was assumed. The "Carbon dioxide, non-fossil" and "Carbon dioxide, in air" flows were thus characterized by a factor equal to 1. In the EF 3.0 methods, methane was also differentiated according to its origin (fossil or biogenic). The corrected characterisation factor for atmospheric methane allowed for the assumption that the carbon in the methane was initially removed from the atmosphere (UNEP-SETAC 2016). It was therefore only required when the atmospheric CO_2 neutrality assumption was applied. The characterisation factor of atmospheric methane is corrected in order to be equal to the characterisation factor of fossil methane.

Two main types of modelling approaches exist in LCA: attributional and consequential. In attributional LCA, the objective is to evaluate the environmental impact that can be associated with a product or service production. It is assumed that the background system is not modified by the studied system (Soimakallio et al. 2015; UNEP-SETAC Life cycle Initiative 2011). In consequential LCA, the objective is to evaluate the environmental impact of changes due to decisions or variations in demand/supply (UNEP-SETAC Life cycle Initiative 2011). The flows modelled in the LCI included all the flows that vary between the baseline scenario and the evaluated scenario. Typically, in attributional LCA the average electricity mix is used for modelling electricity consumption, while in consequential LCA the mode of production affected by the increase in electricity consumption is identified and modelled (marginal mode of electricity production) (Jolliet et al. 2015). Consequential LCA was recommended by Brander et al. (2021), Goglio et al. (2020) and Ramirez et al. (2020) for decision-making purposes. Schaubroeck et al. (2021) advised the choice between attributional and consequential LCA according to the goal of the LCA. Attributional LCA was selected to model the present case study. This choice is discussed in section 4.4.

The calculations were performed with Brightway2 (Mutel 2017). The inventory data and Python script are provided as online resources (see Data availability statement).

3 RESULTS

The results support the discussion on the relevance of methodological choices to perform the LCA of CCUNET systems. They should not be used for deciding whether or not to deploy the CCUNET system described in the case study. In the first subsection, the results related to goal 1 are presented, i.e. on the comparison of a CCUNET system with a conventional counterpart. The second subsection presents the results related to goal 2, i.e. on the potential of the system to generate net negative emissions.

3.1 RESULTS OF GOAL 1 - COMPARISON WITH A CONVENTIONAL COUNTERPART

Two conclusions can be drawn from Figure 12. Firstly, the CCUNET system only performs better than the business as usual situation in the “climate change” impact category. The numerous environmental trade-offs are mainly due to the consumption of heat and hydrogen during i) the transformation of CO_2 into a valuable product and ii) the capture of CO_2 at the end-of-life. It contributes for instance to 20% of the total impact on “energy resources: non-renewable”, 9% of the total impact on “eutrophication” and 10% of the total impact on “particulate matter formation”. However, even when a decarbonized production of heat and hydrogen (amount consumed set to 0 to cancel the impacts) is assumed, environmental trade-offs remain, such as “energy resources: minerals and metals” or “human toxicity, non-carcinogenic, inorganic”. The main contributors are the catalysers used for methanol production and notably copper oxide, but also water production, electricity and sodium hydroxide. This highlights the importance of calculating the impacts not only on climate change but also on all the other impact categories with an exhaustive LCI. Secondly, the CCUNET system is environmentally less beneficial than direct CO_2 storage. When answering the second question, “Is it environmentally better (1) to capture, valorise and then permanently store the CO_2 (CCUNET) or (2) to capture and permanently store the CO_2?”, both the CCUNET system and the reference system aim at generating net negative emissions. Both systems treat 0.6 kg of CO_2 emitted by the CO_2 source. Both systems initially capture 2.2 kg of CO_2 through photosynthesis. The CCUNET system then permanently stores around 0.4 kg of atmospheric CO_2 (treatment at the end-of-life of the CO_2-based product) through geological storage. The reference system stores around 1 kg, i.e. 0.6 kg of atmospheric CO_2 from the CO_2 source and 0.4 kg of fossil CO_2 from the treatment of the fossil-based product. The impact of the CCUNET system on climate change is 3.8 kgCO2eq. The impact of the reference system is 3.1 kgCO2eq. Neither system reaches net negative emissions.

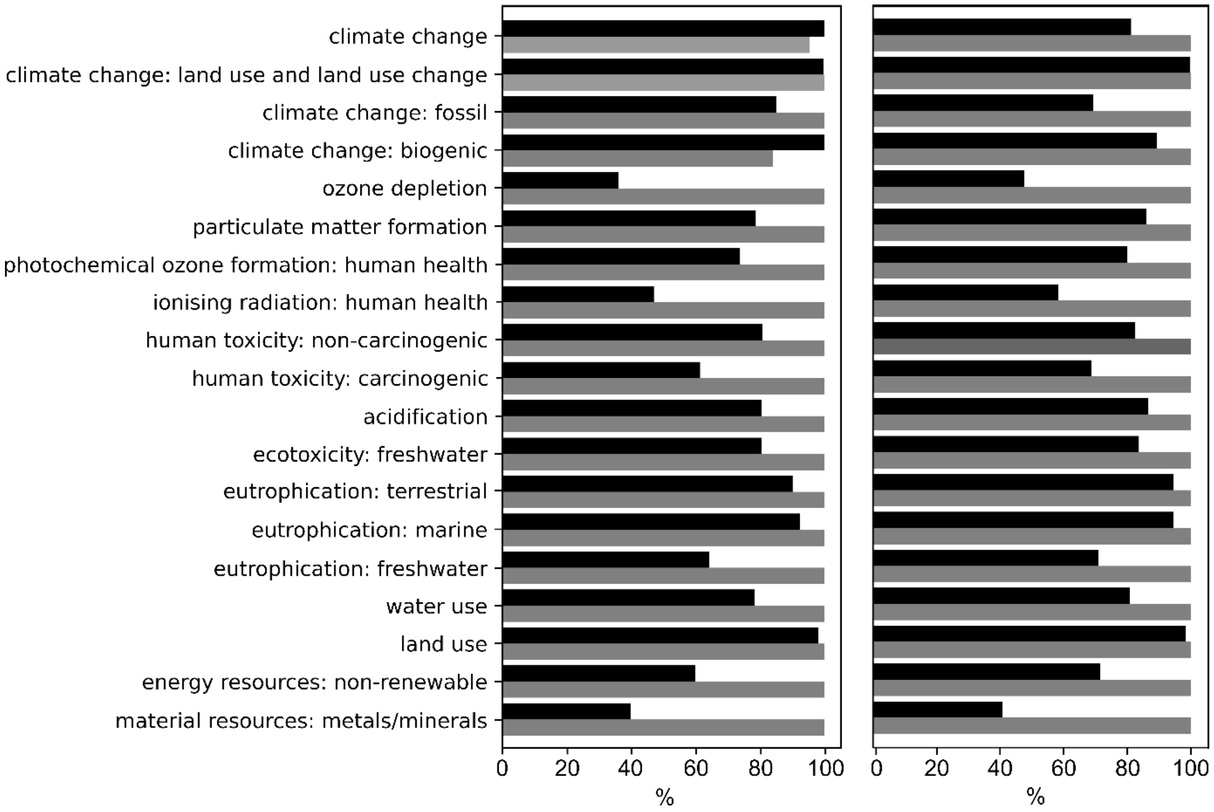

Figure 12: Comparison of the CCUNET system (grey bars) with its conventional counterpart excluding carbon capture (black bars, left-hand graph) and with its conventional counterpart including carbon capture and storage (black bars, right-hand graph). Results were internally normalized with the value of the highest impact obtained in each environmental category

3.2 RESULTS OF GOAL 2 - ARE NET NEGATIVE EMISSIONS GENERATED?

As atmospheric CO_2 is accounted for in both LCI and LCIA (“+1/-1” approach), the biomass production step (1 in Figure 13) generates negative emissions. The amount of CO_2 captured by photosynthesis during biomass growth is not offset by the GHG emissions due to biomass cultivation and harvesting. To better illustrate the methodological issue on multifunctionality, heat and hydrogen were assumed to arise from renewable sources (i.e. impact on climate change = 0). This explains the low contribution to the total impact on climate change of the steps of alcohol fermentation (2 in Figure 13), of the CO_2 conversion to product (3 in Figure 13) and of the CO_2-based product end-of-life (4 in Figure 13). The end-of-life of ethanol and DDGS (5 in Figure 13) has a strong impact on climate change because all the carbon contained in the ethanol and DDGS is released as CO_2 and CH_4 to the atmosphere. Consequently, the impact on climate change of the whole system (Figure 13a) is about 3 kgCO2eq, so no negative emission is reached.

When solving multifunctionality to calculate the impact of producing the CO_2-based product, the end-of-life of ethanol and DDGS were removed from the system boundaries. This is the main reason for the resulting net negative emissions when substitution (Figure 13b) or allocation on carbon content (Figure 13c) are used for solving multifunctionality. When using the circular footprint formula (Figure 13d), CO_2 is considered as a waste. The biomass production and transformation steps are therefore not included in the system boundaries, thus removing the potential for negative emissions. The conclusion on whether the system described in Figure 11 can generate net negative emissions depends on the method selected for solving the multifunctionality. The result from system expansion points out that the system does not generate net negative emissions. However, by using allocation on carbon content (or substitution), an LCA practitioner can claim that the production of the CO_2-based product generates net negative emissions.

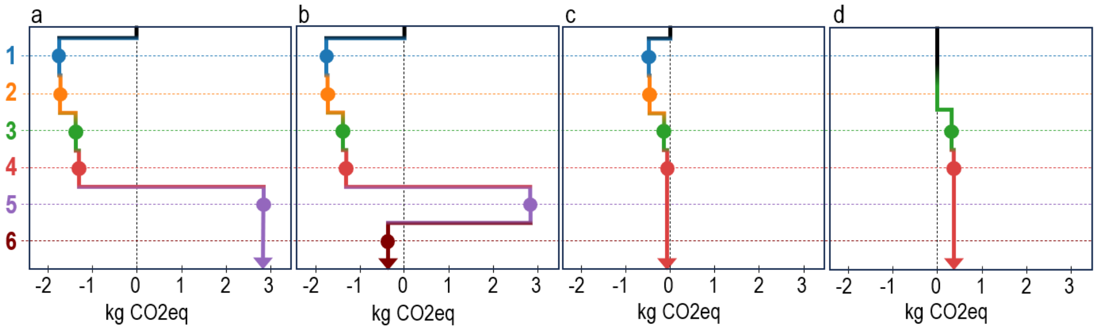

Figure 13: Impact on climate change (Global warming potential - 100 years) according to multifunctionality management. a: System expansion, b: Substitution or allocation by physical causality, c: Allocation on carbon content, d: Circular footprint formula. The impact of each step is added to the previous total impact, starting with 1: Biomass production and followed by 2: CO_2 source, 3: CO_2 to product, 4: End-of-life of the CO_2-based product, 5: End-of-life of the CO_2 source products, and finally ending with 6: Avoided emissions

4 DISCUSSION

In this section, the relevance of applying CCU guidelines to CCUNET systems is discussed relative to each of the two defined goals. Recommendations specific to a given CCUNET system in addition to the existing guidelines for the CCU are formulated when necessary. In the first subsection the relevance of the CCU guidelines for defining the functional unit is examined in the light of existing recommendations for NET systems. In the second subsection, a recommendation specific to CCUNET systems is proposed in order to deal with the multifunctionality of the CO_2 source. In the third subsection, the relevance of the "+1/-1" approach to account for atmospheric CO_2 is discussed. Lastly, in the fourth subsection, attributional LCA versus consequential LCA is discussed for each goal.

4.1 ADDING THE "CARBON REMOVAL" FUNCTION TO THE FUNCTIONAL UNIT?

Jeswani et al. (2022) and Terlouw et al. (2021) highlighted that the primary goal of a NET system is to remove CO_2 from the atmosphere. Therefore, they recommended that the "carbon removal" function should be included in the functional unit for every NET system, whatever the initial goal of the study. However, they observed that such functional units are never used, except in the LCA of systems that are dedicated to the permanent removal of CO_2 from the atmosphere (e.g. direct air capture with carbon capture and storage). This can be explained by the fact that these LCAs do not aim at comparing NET systems between one another. Their goals are rather to compare several production routes in order to improve the environmental performance of the studied systems. The CCS is consequently a means for reducing the system environmental impact on climate change instead of being the main function of the system.

A function such as the "mass of CO_2 stored" is only relevant in the case of a comparative study between NET systems. If the goal were different, the inclusion of this function might not be relevant and could even be misleading. For instance, the system boundaries would be modified if the "mass of CO_2 stored" function were considered for the case study in order to answer the second question, "Is it environmentally better (1) to capture, valorise and then permanently store the CO_2 (CCUNET) or (2) to capture and permanently store the CO_2?". Results indicate that the reference system permanently stored 1 kg of CO_2 compared to 0.4 kg for the CCUNET system. Therefore, for the CCUNET system to be equivalent to the reference system on the "mass of CO_2 stored" function, a system that can store 0.6 kg of CO_2 would have to be included within the system boundaries of the CCUNET system. The extension of the system boundaries in order to include a NET system would lead to a decrease of the impact of the whole system on climate change. However, this decrease remains artificial and only results from the choice of the functional unit.

In the case of a comparative study between NET systems, several reference flows could be used for quantifying the “carbon removal” function; these could be the amount of carbon initially removed from the atmosphere (e.g. by photosynthesis during biomass growth) or the amount of carbon permanently stored at the end of the life cycle. The results from the case study point out a same amount of CO_2 removed by photosynthesis during biomass growth for both the CCUNET system and the reference system. However, the amount of permanently stored atmospheric CO_2 differs by 50%. This confirms the importance of a precise definition of the functional unit and of the associated reference flow.

Jeswani et al. (2021) proposed a functional unit of “1 t of CO_2 (sequestered, removed and/or stored)”, which points to an ambiguity in the definition. Zakrisson et al. (2023) proposed “Carbon dioxide sequestered”, corresponding to the amount of CO_2 stored in the long-term storage. Goglio et al. (2020) used “CO2eq removed” as an example. Terlouw et al. (2021) interpreted the use of “CO2eq” as the carbon removed from the atmosphere, calculated over the whole life cycle using LCA. It is the result of the impact assessment on climate change. This implies the necessity to carry out an LCA of the system before the functional unit can be defined, which is not very practical. It also suggests that the functional unit depends on the characterisation factors used to calculate the impact, thus adding uncertainties. Zimmermann et al. (2020) proposed to use “mass of CO_2 utilized” as a functional unit to compare CCU and CCS systems. Drawing on this proposal, the “treatment of atmospheric CO_2” function could be added to the functional unit of CCU or NET systems. Treating atmospheric CO_2 refers to any capture of atmospheric CO_2 (from the atmosphere or from an industry based on biomass) as well as its subsequent treatment (valorisation, storage…).

In the case study, this would imply the addition of the “treat 0.6 kg of atmospheric CO_2” function. It would not modify the results if the release of CO_2 to the atmosphere were considered as a sort of treatment. With this approach, “treating atmospheric CO_2” may or may not entail net negative emissions. If net negative emissions were indeed achieved, the impact in the ‘climate change’ category would represent the efficacy of the process (i.e. the amount of net negative emissions generated by mass of treated atmospheric CO_2). The impacts in the other categories could be normalized by the impact on climate change at a later stage if the results on mass of removed CO_2eq are required. The vocabulary “remove CO_2 from the atmosphere” (carbon sequestration, e.g. Gt or Mg of CO_2eq removed) could then be employed exclusively to qualify the expected or true result of the system in the climate change impact category. Whatever functional unit is chosen to include carbon removal, a comparative study between NET systems requires multifunctionality to be dealt with, since only direct air capture with carbon capture and storage or enhanced weathering are monofunctional. This topic is covered in the following subsection.

4.2 RECOMMENDATION TO SOLVE MULTIFUNCTIONALITY FOR EVALUATION OF NEGATIVE EMISSIONS

With system expansion, all the carbon captured initially from the atmosphere by the system is tracked until its reemission to the atmosphere or its permanent storage. Thus, with system expansion, a negative value obtained in the "climate change" impact category can be interpreted as net negative emission. However, the results indicate that, in contradiction with Terlouw et al. (2021) (cf. Table 1), an allocation based on physical criteria does not guarantee that a negative value obtained in the "climate change" impact category could be interpreted as net negative emission. In the case study, a negative value was obtained when the allocation on carbon content was used for solving multifunctionality. However, it should never be used as background inventory data to offset GHG emissions in another life cycle, because this would overestimate the mitigation potential of the product. Indeed, if the whole production system did not reach net negative emissions, an increase in the production of the product for which net negative emissions are allocated would not lead to less CO_2 in the atmosphere. The net negative emissions allocated to a given product would only offset the GHG emissions allocated to the other co-products of the system (e.g. the emissions due to the end-of-life of ethanol and DDGS). Only system expansion is compatible with evaluating the potential for net negative emissions.

If multifunctionality needs to be solved, system expansion should be applied first in order to verify whether the whole system reaches net negative emissions. Then, in order to avoid the overestimation of the mitigation potential of the products, the method selected for solving multifunctionality should not entail negative results for any of the products if the whole system cannot achieve net negative emissions. When focus is put on climate change, a solution could be to assume that the production of all products is climate neutral, in agreement with the goal of achieving carbon neutrality by 2050. The net negative emissions achieved would then be entirely allocated to the "treatment of atmospheric CO_2" function (cf. previous section). In fact, the net negative emissions calculated with system expansion can be used for offsetting the impact on climate change of another system. However, this approach does not allow for the allocation of the impact to other impact categories. Moreover, there is a risk that solutions which maximise net negative emissions rather than net zero products could be preferred to solutions which maximise the production of net zero products. Consequently, carbon accounting the net negative emissions for legal purposes, such as product declaration, deserves further investigation towards a solution that does not overestimate the environmental benefits of the products.

4.3 RELEVANCE OF THE "+1/-1" APPROACH TO ACCOUNT FOR BIOGENIC CARBON

The "+1/-1" approach was applied to answer both goals, even though it was not specifically recommended by existing guidelines. The main recommendation for atmospheric CO_2 accounting is to include it in the LCI but also to assume that it does not produce an impact on climate change (characterisation factor equal to zero, see Table 1). Nevertheless, if the impact of atmospheric CO_2 were counted as zero, a positive effect on climate change due to atmospheric CO_2 permanent storage would still need to be included so as to avoid overestimating the impact. Masses of captured CO_2 are included in the inventory as negative amounts, i.e. negative flows. A characterisation factor of 1 is then applied, leading to a positive effect on climate change. Negative flows are usually added at the stage of CO_2 permanent storage or at the step of technological CO_2 capture from industrial fumes. Although mathematically correct, this approach may cause methodological misinterpretation. Indeed, positioning the benefit of atmospheric CO_2 capture at the technological capture stage could suggest that the same credit can be given to fossil CO_2. Positioning CO_2 capture at the stage when it physically takes place, e.g. CO_2 captured by photosynthesis during biomass growth, reduces this risk of misinterpretation. In addition, it allows for atmospheric and fossil CO_2 emissions to be treated identically, which is more coherent since they concern the same molecule. Lastly, thanks to the "+1/-1" approach, a dynamic impact assessment can be performed.

The construction of the inventory can become more complex with the "+1/-1" approach, particularly for the step related to food or feed. The end-of-life of DDGS is one of the main contributors to the impact on climate change in the case study. Therefore, to achieve good quality estimates of net negative emissions, further research is necessary in order to build inventory data on the tracking of the fate of carbon until its re-emission to the atmosphere. This requires the establishment of conversion factors from the mass of ingested carbon to the mass of emitted CO_2 or CH_4, or to a lesser extent, ranges of values allowing for a significant sensitivity analysis to be performed.

Concerning the impact characterisation step, a symmetry in the response of the climate system between CO_2 emission and capture was assumed. Zickfeld et al. (2021) obtained an asymmetry in the climate model response for a peak emission of -100 $GtCO_2$. Therefore, one hundred years after the negative emission peak, 51 $GtCO_2$ would still be fixed outside the atmosphere. In the case of an emission peak of the same magnitude, one hundred years later, 53 $GtCO_2$ would still be present in the atmosphere. This difference is small compared to the uncertainties presented by Joos et al. (2013). Joos et al. (2013) reviewed the results obtained from 16 climate models, including the one used by Zickfeld et al. (2021). Joos et al. (2013) observed that after one hundred years 52.4 ± 11.3 $GtCO_2$ would still remain. It would be therefore be noteworthy to repeat a similar investigation on peak captures of

-100 $GtCO_2$ in order to determine whether the asymmetry noted by Zickfeld et al. (2021) is significant and well reflected in all other models.

4.4 ATTRIBUTION LCA OR CONSEQUENTIAL LCA FOR EVALUATION OF NET NEGATIVE EMISSIONS?

In the following subsections, attributional versus consequential LCA is discussed relative to the two defined goals.

4.4.1 Goal 1 - Comparison with a conventional counterpart?

In section 2.1, this goal was split in two to compare the CCUNET system with a business as usual situation and with a system based on direct permanent storage of captured CO_2. These can be interpreted as a consequential question: “What are the environmental consequences of using a CO_2 treatment solution (CCS or CCU)?”. The application of consequential modelling rather than attributional modelling would lead to the following changes in the construction of the inventory.

Firstly the background data would be modified. The production of electricity, heat and hydrogen (main contributors to the impacts of CCU systems as highlighted by the results) are unconstrained markets. Their production would therefore be modelled with marginal data rather than with averages. Due to the increasing demand, the marginal energy mix often involves more recent technologies than the average energy mix, (Weidema 2003). The marginal energy mix therefore represents a more optimistic scenario than the average energy mix. The production of raw materials, such as catalysers, can be provided by constrained markets, i.e. the material would only be produced as a by-product of a given process. In this case, with consequential LCA, an increase in the demand for such a type of material (noted 1) would not cause an increase in production but rather the substitution of material 1 by another (noted material 2) from a process where this is possible. It is the increased production of material 2 that would then be modelled in order to calculate the impact of an increase in demand for material 1. Consequential modelling can lead to different results than attributional modelling.

Secondly, the foreground data would also change, because the consequences of implementing the system on the market would also need to be analysed. Would the production of propylene from CO_2 lead to a reduction in the production of propylene from fossil sources, or would it meet an increase in demand? Would the cost of adding carbon capture to an ethanol plant increase the price of ethanol and subsequently reduce the demand? Or would subsidies encourage CO_2 storage, resulting in an increase in CO_2 production and therefore an increase in ethanol production, biomass use and, ultimately, changes in land use? As noted by Goglio et al. (2020), proposing the answers to these questions is currently challenging. Mechanisms, such as government policies or social acceptance, that influence the deployment of CCU or CCS are still being developed or still need to be understood and measured. Models, e.g. economic or agent-based, integrating such mechanisms should first be developed before these questions can be answered and consequential LCA can be properly applied.

Attributional LCA do not take into account the consequences that are not physically related to the systems (e.g. market effect). However, attributional LCA is a tool for beginning to sort out existing solutions. Attributional LCA comparison can be made in order to identify burden-shifting and the maximum benefit that could be obtained. The maximum benefit is achieved when a perfect substitution can be made between a new production method and an old one (Plevin et al. 2014). If an attributional LCA comparison were not conclusive, there would not be a need for further study. Consequential LCA would be applied when there is a better understanding of the markets in order to verify that solutions that have not been rejected by attributional LCA would be interesting for large-scale deployment and for constituting relevant public policies. Consequential LCA could therefore contribute to optimise an economic model under environmental constraints in order to assess the types of public policies needed for achieving the environmental objectives.

4.4.2 Goal 2 – Are net negative emissions generated?

This goal falls within the definition of an attributional goal, i.e. "gaining a qualitative understanding of a production system" (Plevin et al. 2014). Furthermore, consequential LCA can produce negative result values, due to avoided emissions (Schaubroeck et al. 2021). The inability of LCA calculation software to distinguish between avoided emissions and negative emissions represents a barrier for calculating net negative emissions. In addition, when the ultimate goal is to assess whether net zero emissions can be achieved, the whole actual system, and not just the marginal parts, would have to be evaluated. In conclusion, attributional LCA appears to be compatible with the assessment of the potential for net negative emissions.

5 RECOMMENDATIONS

Results indicated that CCU guidelines are not completely compatible with the evaluation of CCU systems designed to generate negative emissions. Three recommendations specific to the evaluation of CCUNET systems, and related gaps in the current scientific literature, have emerged from the discussion, and should be used in addition to the existing CCU guidelines:

- Fully account atmospheric carbon in the LCI and in the LCIA. Further research should focus on two points. First, knowledge on the fate of carbon during food and feed steps needs to be improved. Second, the assumption of symmetry in the response of the climate system between CO_2 emissions and capture should be verified. If it is not valid, a specific characterisation factor for atmospheric CO_2 capture must be calculated.

- To compare CCUNET systems to other NET systems, use the “treatment of atmospheric CO_2” functional unit as the common function between systems and solve multifunctionality with system expansion.
- To calculate the net negative emission potential, use attributional LCA, with “from-cradle-to-grave” system boundaries as well as system expansion to solve multifunctionality. Further research must be carried out to solve multifunctionality for carbon accounting of net negative emissions (e.g. product declaration) that does not overestimate the environmental benefits of the products.

6 DATA AVAILABILITY STATEMENT

The inventory data used to perform the LCA of the case study is provided in the Excel® file ESM1. The conda environment to run the scripts and perform LCA with Brigthway v2 is detailed in the yml file ESM2. The Brightway2 database was created by importing the Excel® file and slightly modifying it with the jupyter notebook ESM3. The data illustrated in Figure 12 is generated with the jupyter notebook ESM4. The data illustrated in Figure 13 is generated with the jupyter notebook ESM5. In both cases, the data was reorganised using powerpoint and inkscape in order to produce the figures displayed in the article.

CHAPTER DISCUSSION: LIMIT OF THE CHOSEN CASE STUDY

Two important choices were made in the definition of the case study: land use change (LUC), that could occurred upstream of the biomass production, was not included and only incineration is considered as end-of-life of the CO_2-based product. As stated in the introduction the CO_2-based product could be landfilled or recycled for generating negative emissions (Figure 1). In the following subsections, it is shown that including LUC (first subsection), and studying these two alternative end-of-life (second subsection) raises further methodological challenges for evaluating the negative emission potential of CCU systems.

LAND USE CHANGE

LUC can be direct or indirect. Direct LUC occurs when the function of a land is changed, e.g. from forest to cropland. This change generally leads to emissions of biogenic carbon due to the destruction of the previous biomass (both above and below ground) and machine use. Indirect LUC occurs when the previous production is displaced leading to direct cascading LUC. For example, transforming cropland for food into cropland for bioenergy is not considered as LUC if the type of crops produced on the land does not change. However, in the case of constant food demand, food still needs to be produced elsewhere and can generate direct LUC on another land (e.g. forest to cropland). This direct LUC for food is an indirect LUC linked with the production of bioenergy (Brandão et al. 2022). Indirect LUC is thus included in consequential LCA rather than in attributional LCA, recommended to evaluate negative emissions potential in this chapter.

Direct LUC can be both included in the LCI and characterized as an impact. Bhan et al. (2021) reviewed methods to quantify land-use-induced carbon emissions. They distinguish two approaches: "actual forestry and other land use emissions" and "sequestration potential foregone"; the former enables to include LUC in the LCI in attributional LCA. The difference between the carbon stocks of the actual land use and the carbon stocks of the previous land use is calculated to evaluate the flows of biogenic carbon due to LUC. It corresponds to the approach used for national GHG inventories. Goglio et al. (2015) found that the IPCC Tier 1 and Tier 2 methods (IPCC) are recognised, accepted and applicable methods to include LUC in LCA. However, this "actual forestry and other land use emissions" approach can mask the impact of previous deforestation. For instance if deforestation is followed by annual crops plantation during five years and then permanent crops plantation, the impact of deforestation is allocated only to the first use. The second approach, "sequestration potential foregone", is rather used to characterize the impact of LUC (Müller-Wenk and Brandão 2010; UNEP-SETAC 2019). The aim is to calculate the amount of carbon sequestration that would have occurred in the defined area with the reference land use. The flows characterised describe the land occupation (m^2.yr) and the land transformation (m^2) (UNEP-SETAC 2019). This "sequestration potential foregone" can also be used in consequential LCA to include LUC in the LCI. One major challenge is the definition of the land reference. Koponen et al. (2018) propose a framework to help choose the land reference between "no-human intervention" or "most likely other land use" depending on the goal of the LCA. The "sequestration potential foregone" approach is less developed than the "actual forestry and other land use emissions" approach notably because the carbon stocks in the reference land use are hypothetical and uncertain (Bhan et al. 2021).

Research on LUC and foregone sequestration notably arise from critics of bioenergy impact assessment on climate change. Bioenergy was developed to reduce the climate change impact of energy consumption in transport, residential-tertiary or industrial sectors. The main asset of bioenergy is the assumption of biogenic CO_2 neutrality on climate change. To evaluate without bias the mitigation potential of bioenergy, Brandão et al. (2013) propose to keep the biogenic CO_2 climate neutrality assumption, to add in the inventory all the carbon emissions or captures due to SOC and LUC changes, and to compare the bioenergy system to a reference product system whose energy production is from fossil sources. Such reference system must be distinguished from the land reference used to calculate

LUC. When the bioenergy system is responsible for LUC (direct or indirect), Cherubini et al. (2009) indicate that the reference system should also include an alternative land or biomass use. However, in an attributional LCA of a CCUNET system, land use is not a function or a reference flow of the system but rather an impact to characterise.

Therefore, our recommendation is to not include an alternative land or biomass use in the reference system. LUC should be modelled by two types of flows:

i. Occupation and transformation flows, that will be used to calculate the impact of the system in the impact category “land use”. It enables to include the effect of LUC as a characterized impact on soil quality, biodiversity, foregone sequestration and so on.
ii. Pollutants flows such as greenhouses gases (CO_2, CH_4…) due to direct land transformation (machine use, biomass destruction). It enables to include the immediate effect of LUC in existing impact category such as climate change.

END-OF-LIFE

Landfill is considered for long term storage of carbon because of the assumption that plastic takes hundreds of years to decay. However, Chamas et al. (2020) observed a lack of knowledge on the degradation rate of plastics. They point out that the degradation times quoted in the media vary from dozens to thousands of years without any solid scientific evidence. They observed that in the current scientific literature some of the information needed to correctly interpret the results is not provided, such as the factors that can accelerate degradation (temperature, humidity, size of waste). In addition, extrapolation methods are used to estimate plastic lifetime, as it would otherwise require experiments lasting several decades. Depending on the method chosen, it can lead to discrepancies of several decades or even hundreds of years. If the lifespan of plastics in landfill sites is indeed of several hundred years, landfill is a possibility for permanent carbon storage. This solution may seem interesting when looking at the LCA results for negative emissions production. However, LCA does not robustly evaluate the impact of microplastic pollution. For example, groundwater may be polluted by microplastics from landfill sites (Manikanda Bharath et al. 2021). Jiao et al. (2024) reviewed the research gaps linked to LCA of plastic waste management. They conclude that there is a lack of knowledge about the impact of microplastics on human health and ecosystems. Moreover, the European Union is aiming to limit the proportion of municipal solid waste treated in landfill to just 10% by 2035 (European Commission 2018). Landfill is the least preferable option on the European Union’s waste hierarchy both for pollution reasons (groundwater contamination and methane production) and for circular economy reasons (burying recyclable materials is a waste of materials).

Plastics produced from CO_2 could be recycled at their end-of-life through two main recycling processes: close-loop and open-loop. For both, a fraction of the recycled product is converted into new raw material and a fraction is converted into non-recoverable waste. The difference lies in the use of the new raw material. In close-loop recycling, the new raw material is of similar quality than the pre-recycled material and is used for producing the same type of product. In open-loop recycling, the new raw material can have very different properties from the pre-recycled material and is used for an entirely different purpose. Recycling is a multifunctional process shared between two lifecycles. It has both the function of treating waste material and of producing new raw materials. Following the recommendations proposed in this chapter, system expansion needs to be used to deal with this multifunctionality. For open-loop recycling, defining the system boundaries is challenging, as system expansion implies including in the system boundaries all the successive lifecycles of the material. For instance, if the recycled plastic is used to produce a car-bumper, how much of the car lifecycle is it relevant to include in the system boundaries to evaluate if the CCUNET system generates negative emissions? For close-loop recycling, the recycled share of material is noted R. The impact of production in close-loop recycling of unit of product can then be calculated as:

$$I_{close-loop\ recycling} = (1-R)I_{primary\,production} + R \times I_{recycling} + (1-R)I_{CCS} \tag{1}$$

With:

- $I_{primary\,production} + I_{CCS}$ is the impact of the production of the CO_2-based product and its end-of-life (incineration with carbon capture and storage). If the primary production system effectively generates negative emissions, then $I_{primary\,production} + I_{CCS} < 0$.
- $I_{recycling}$ is the impact of the recycling process.

The case $R = 0$ corresponds to no recycling. The whole system generates negative emissions only if $I_{recycling} < -\frac{(1-R)}{R}\left(I_{primary\,production} + I_{CCS}\right)$. So either the impact of the recycling process is small enough to be offset by the negative emissions generated by primary production system or the recycling process generates negative emissions, for instance by using BECCS to produce the needed energy. If the goal is to generate negative emissions, the more primary material is produced and treated, the more CO_2 is removed from the atmosphere. The recycling process reduces the potential of the system to produce negative emissions, unless the recycling process generates more negative emissions than the primary production process per unit of material produced. However, recycling can have a positive influence in other impact categories.

In conclusion, evaluating the potential of negative emission of systems including landfill is limited by a lack of knowledge regarding the degradation of plastic in landfill and about the impact of microplastics on human health and ecosystems. Evaluating the potential of negative emission of systems including close-loop recycling does not raise new methodological challenges. A right amount of recycling can enable to find a balance between generating negative emissions and limiting impact transfers. However, evaluating open-loop recycling will be challenging to define relevant system boundaries with system expansion.

CHAPTER CONCLUSION

In this article, recommendations specific to the evaluation of CCUNET systems are formulated to be used in addition to the existing CCU guidelines. A point is left for future study: the timing of removal and emissions of atmospheric CO_2. This article demonstrates that the "+1/-1" approach is usable, despite some lack in LCI data for the food and feed steps. To tackle the issue of the timing of removal and emissions of atmospheric CO_2, temporal information can be added to the LCI containing a full accounting of atmospheric CO_2 in order to carry out a dynamic LCA. This topic is covered in the following chapter. Dynamic LCA is performed only for the impact category "climate change". However, it should be noted that, as this article illustrates, CCUNET systems can be relevant in terms of climate change, but can also generate significant impact transfers.

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Chapter 3: Facilitating dynamic life cycle assessment for climate change mitigation

This chapter is ready to be submitted to the journal ‘Sustainable Production and Consumption’: Duval-Dachary S., Lorne D., Batôt G., Helias A., Facilitating dynamic life cycle assessment for climate change mitigation. Calculating the impact of biomass use on climate change is still controversial, notably in terms of how to account for differences in the dynamics of carbon storage by photosynthesis and releases at the end-of-life. Applying dynamic life cycle assessment (LCA) is the appropriate answer, but it requires more data and increases the complexity of the calculation. The aim of this article is to explore how to make dynamic LCA easier to use in this context, through a case study. Three purposes are addressed, the modelling tool, the time dimension in the functional unit and the contribution of the time dimension to the accuracy of results.

List of abbreviations

(A)GWP	(Absolute) Global Warming Potential
BECCS	BioEnergy with Carbon Capture and Storage
CCS	Carbon Capture and Storage
CCU	Carbon Capture and Utilisation
EP_{conv}	Scenario with conventional energy production (heat and hydrogen production modelled by Ecoinvent datasets)
EP_{zero}	Scenario with perfectly decarbonised energy production (heat and hydrogen is set to zero)
FU	Functional Unit
FU_1	Production of 20000 bags over the entire lifespan of the plant (LP)
FU_2	Production of 20000 bags at t_0
GTP	Global Temperature Change Potential
IPCC	Intergovernmental Panel on Climate Change
LB	lifespan of the bag
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
NET	Negative Emission Technology
SI	Supplementary information
SOC	Soil Organic Carbon
SOC_{high}	Scenario where miscanthus production leads to soil organic carbon stock decrease
SOC_{low}	Scenario where miscanthus production leads to soil organic carbon stock increase

List of the main variables

LP	Lifespan of the plant (duration, years)
P_0	First year of production (date, year)
P_{end}	Last year of production (date, year)
TH	Time horizon (duration, years)
t_0	First year of the time horizon (date, year)
t_e	time of emission or capture of a greenhouse gas (date, year)

CHAPTER CONTEXT

The scientific literature agrees on the importance of including in the evaluation of the dynamic of uptake and release of atmospheric CO_2, not only for NETs but also for CCU systems (Bui et al. 2018; Goglio et al. 2020). Dynamic LCA, presented in section 4.3 of the introduction, is the solution, but currently too complicated for widespread use. This chapter aims at contributing to the simplification of dynamic LCA through improvement of the existing tool and recommendation on key methodological issues. The first subsection explains why a specific tool is needed and presents the existing tools. Moreover, in chapter 1, we showed that the quality of inventory data also has a considerable impact on the results. A LCA practitioner usually have a limited time to carry out an LCA and needs to find the right balance between additional effort and additional accuracy of the results. The last point of this chapter is to explore through a case study if the variation in results induced by dynamic characterisation of the impact on climate change is significant compared with the variations induced by uncertainty on inventory data. The second subsection presents the specificity of the case study used to illustrate the methodological issues raised in this chapter.

MODELLING TOOL

In static LCA, the calculation of the LCI is based on matrix computation, following the formulas mentioned by Heijungs and Suh (2002):

$$s = A^{-1}f \tag{1}$$

$$inventory = Bs \tag{2}$$

With:

- f: FU vector. Each line represents a product (material, energy, service). The values given in the matrix represent the functional unit of the studied system (ex: 1 for the line 'CO_2-based bag' and 0 elsewhere).
- A: technological matrix. Each line represents a product (material, energy, service). Each row describes an independent unit process (single-product output). Thus, each row contains the amounts of materials, energy or services consumed to produce one unit of product. The matrix A is square.
- s: scaling vector. Each line represents an independent unit process. The scaling vector contains the amounts of each independent unit process necessary to produce the functional unit.
- B: matrix of environmental interventions. Each line represents an elementary flow (emissions or resources). Each row describes an independent unit process (single-product output). Thus,

each row contains the amount of substance directly emitted (or direct resource consumption) during the process to produce one unit of product.

Therefore, the system is divided in independent unit processes. It is possible to provide temporal distribution in the description of each independent unit process, at least of the foreground system. The foreground system “consist of processes which are under the control of the decision-maker for which an LCA is carried out” (Frischknecht 1998). The foreground system is completed by the background system, which is not, or only indirectly, under the control of the decision-maker.

The challenge is computational: how to propagate time distributions between unit processes? Beloin-Saint-Pierre et al. (2014) proposed first a solution: i) provide discrete temporal distribution for each input, output or environmental intervention and ii) propagate time distributions thanks to product of convolution. Then Tiruta-Barna et al. (2016) proposed to use continuous temporal distributions. They found that product of convolution unexpectedly deforms the shape of continuous temporal distributions. Thus, product of convolution cannot be used to properly propagate continuous temporal distributions. Instead they propose to look at the LCI as a network directed graph (node = process, arc = exchange). A graph traversal algorithm can then be used to solve the system. Tiruta-Barna et al. (2016) also propose to add an information, the supply model. The supply model is an additional temporal distribution that describes the acquisition of the product. Thus, the supply model includes information such as the delay due to storage or transport. This methodology has been operationalized in a web tool, DyPLCA (Pigné et al. 2020). Cardellini et al. (2018) operationalized the method proposed by Beloin-Saint-Pierre et al. (2014) (Temporalis package, Brightway2).

In this work, Temporalis was selected rather than DyPLCA for several reasons:

- The focus of this work is the dynamic assessment of the impact on climate change. A time step of a year is sufficient. Continuous temporal distributions are thus unnecessary.
- Temporalis is part of Brightway2. It enables to use the same tool to perform dynamic and multi-criteria static LCA.
- Temporalis is written in the Python programming language and is open-source. It is thus transparent and modifiable.

CASE STUDY FOR DYNAMIC LCA

Temporally distributed background databases do not exist yet. Thus, in this work temporal information will only be added in the foreground system.

The case study used in the previous chapter is not suitable to illustrate dynamic LCA issues as maize grows in only one year. Thus, the biomass is changed to miscanthus and wood residues, two lignocellulosic feedstocks with different growth temporal pattern. Miscanthus is a perennial crop (not replanted each year) and thus able to generate carbon storage in the soil due to the growth of its root network (see Annexe 2). Wood residues are considered as waste to be treated (i.e. zero-burden product as the impacts were allected to the wood). When they are generated during plant growth or harvest, they can be for example left on the field and contribute to an increase in SOC. They can also be generated in post-harvest processes (e.g. sawdust). Residues (i.e. waste) are allocated emissions and resource consumption only for the steps needed for their transformation into valuable products (e.g. collection, drying, etc) (Nessi et al. 2021). Due to this allocation, the results cannot be used to conclude on the potential of this system to generate negative emissions. The results obtained in this chapter can only be used to illustrate dynamic LCA methodological challenges.

1 INTRODUCTION

The 28th Conference of the Parties held in Dubai reaffirmed the urgent need for action to limit global warming to 1.5°C (European Council 2023). In the Intergovernmental Panel on Climate Change (IPCC) report on global warming of 1.5°C (IPCC 2018), bioenergy with carbon capture and storage (BECCS) is part of the strategy, with an average deployment rate ranging, depending on the scenario, from 3 to 7 $GtCO_2$ per year by 2050. BECCS refer to systems that convert biomass into energy and capture the released CO_2 in order to store it permanently outside of the atmosphere. BECCS generate a flow of CO_2 from the atmosphere (capture by photosynthesis during biomass growth) to a permanent storage outside the atmosphere (CCS). If the beneficial impact of capturing CO_2 from the atmosphere is not offset by the impact due to greenhouse gas emissions over the entire life cycle of the BECCS system, then the BECCS system generates negative emissions. The mitigation potential of BECCS needs to be assessed. This is addressed using Life Cycle Assessment (LCA) (ISO 2006a; ISO 2006b) to take into account all emissions due to the consumption of energy (e.g. heat for carbon capture) and chemicals (e.g. solvent for carbon capture). There is ongoing research on how to account for the impact of biomass use on climate change (Brandão et al. 2019; Brandão et al. 2024), and the assessment of negative emissions (Brander et al. 2021; Goglio et al. 2020). A key question is how to account for the differences in the dynamics of carbon storage and release and its impact on climate change (Brandão et al. 2019; Brander et al. 2021; Goglio et al. 2020; Jeswani et al. 2022).

Dynamic Life Cycle Impact Assessment (LCIA) is an answer to this problem (Brandão et al. 2024; Brander et al. 2021). The glossary proposed by Beloin-Saint-Pierre at al. (2020) is used as a reference in the present article for vocabulary linked to dynamic LCA (e.g. temporal scope, time horizon and so on). The usual way of performing LCA is referred to as static LCA. Dynamic LCIA is defined as "characterisation models of environmental mechanisms that account for the dynamic of ecosphere systems and can therefore use temporal information of dynamic Life Cycle Inventories (LCI)" (Beloin-Saint-Pierre et al. 2020). The original dynamic LCIA method was developed by Levasseur et al. (2010) to characterise the impact on climate change. For an emission of a greenhouse gas at time t and an impact assessed over a time horizon TH, corresponding to the time between t_0 and t_{end}, Levasseur et al. (2010) proposed to calculate the Absolute Global Warming Potential (AGWP) at t_{end} as the integral of the radiative forcing between t and t_{end}. Dynamic LCIA on climate change is an active area of research, with new characterisation methods (based on GWP (Ventura 2022)) and decision-support indicators (based on Global Temperature Change, GTP (Tiruta-Barna 2021)) continuing to be developed. However, Beloin-Saint-Pierre et al. (2020) point out that carrying out a dynamic LCIA requires significant additional effort, increasing data requirements and the complexity of calculating the inventory. Su et al. (2021) noted a lack of tested tools for calculating both inventory and impact. Brandão et al. (2024) rated the ease of application of dynamic LCIA as rather poor (3/5, 1 being really easy to use). In this context, the aim of this paper is to explore how to make dynamic LCIA easier to use.

2 LITERATURE REVIEW

Firstly, temporal differentiation of the LCI, i.e. distributing on a time scale the consumption and production of each process included in a life cycle, is complex. It is not possible to use conventional LCA software such as Simapro® or Gabi®. Only the open-source python library Temporalis (Cardellini et al. 2018) can be used for calculating a dynamic LCI and then perform a dynamic LCIA. In the present article, dynamic LCI refers to "LCI that is calculated from supply and value chains where […] temporal differentiation is considered resulting in temporal distributions to describe elementary flows" (Beloin-Saint-Pierre et al. 2020). However, as pointed out by Su et al. (2021) and Beloin-Saint-Pierre et al. (2020), Temporalis still needs to be tested to validate its operability and efficiency. Another challenge is the availability of generic dynamic LCIs. Some studies (e.g. (Jury et al. 2022; Zieger et al. 2020)) provide inventory data over the entire lifespan of the system, i.e. for the production of several units of the product or service each year over the entire lifespan of the system. To reuse the data in a different life cycle, it is easier to use an average dynamic LCI, i.e. for the production of one unit of the product or service at time $t_{0,process}$. Defining a process-relative "time 0" ($t_{0,process}$) enables to create process-relative temporal distribution. Testing Temporalis and proposing an algorithm for averaging a dynamic LCI is the first objective of this study.

Secondly, Su et al. (2021) point out that many dynamic LCIA studies compare results obtained using static LCA and results obtained using dynamic LCA. Two types of functional units are observed: i) the production of several units of the product or service each year over the entire lifespan of the system (e.g. (Shen et al. 2022; Zieger et al. 2020)) and ii) the production of one unit of the product or service at t_0 (e.g. (Almeida et al. 2015; Wang et al. 2022)). Using static LCA, the results obtained using the two types of functional units are equal if the total quantity produced is equal. Is the same true when using the dynamic LCA method? Furthermore, in static LCA, the potential impact of the system studied on climate change is generally provided for a single time horizon (usually 100 years) which is not calendar based. Using the dynamic LCIA approach, the potential climate change impact of the system under study is provided for a series of t_{end} defined relatively to a t_0 equal to zero (e.g. (Zieger et al. 2020)) or based on a calendar (e.g. (Shen et al. 2022)). There is currently no consensus on how to position the temporal distribution describing the inventory relatively to t_0. In the work of Negishi et al. (2019) or Almeida et al. (2015), the first year of production is chosen as equal to t_0. It is rather the year in which the infrastructure is built that is chosen as t_0 in the work of Zieger et al. (2020). Ventura (2022) offers yet another perspective by defining a total observation duration corresponding to the sum of the duration of the inventory and the time horizon, which is equivalent to choosing the last year of the inventory as t_0. In this present article, the recommendation of Beloin-Saint-Pierre et al. (2020) is followed, i.e. t_0 is equal to the time when the product, service or system is ready to be used. However, an ambiguity remains when the production occurs over several years. Therefore, the second objective of this paper is to explore the influence of the definition of the functional unit using a case study, in order to propose recommendations for facilitating future interpretation and comparison of dynamic LCA studies.

Thirdly, dynamic LCA results are compared to static LCA results for evaluating if it is worthwhile to perform a dynamic LCA. For example, Almeida et al. (2015) concluded that it was not worth the effort required to perform dynamic LCA. Pigné et al. (2020) added temporal information to a whole background database and observed significant differences only when the datasets included high upstream emissions (due to infrastructure construction). The balance between the complexity of the approach and the addition of precision to the results is thus central to dynamic LCIA. Collet et al. (2014) suggest adding temporal information only to the main contributors to the impact, and only if their temporal scope is equal to or greater than the temporal resolution of the impact, i.e. one year for climate change. Following this recommendation, the third objective of this work is to explore if the variation in results induced by dynamic characterisation of the impact on climate change is significant compared with the variations induced by uncertainty on inventory data.

The purpose of this article is thus to investigate the value and feasibility of explicitly including time in environmental assessments of climate mitigation solutions. An illustrative case study (a reusable polypropylene shopping bag) is used for fulfilling the three underlying objectives described above: testing temporalis, the time dimension of the functional unit and the contribution of the time dimension to the accuracy of results.

3 MATERIAL AND METHODS

The inventory data used for modelling the case study, the method applied for averaging a dynamic LCI and the method to perform a dynamic LCA on climate change are presented in section 3.1. Changes made to Temporalis are described in the section 3.2. The sensitivity analysis performed on the definition of the functional unit and time horizon is presented in the section 3.3. The sensitivity analysis performed to compare dynamic and static LCA is presented in section 3.4.

3.1 CASE STUDY, INVENTORY MODELLING AND DYNAMIC LCIA

The production of a reusable shopping bag from CO_2 was chosen as a case study for its temporal parameters (duration of biomass growth, lifespan of the plant producing the shopping bag, lifespan of the shopping bag) and the availability of inventory data in the literature. The case study is illustrated in Figure 14. Two biomass productions are studied : miscanthus, a fast dedicated production system and wood residues long-term sub-product production system. The biomass is then transformed by alcoholic fermentation into ethanol, electricity and CO_2. The CO_2 is converted to methanol, and then propylene to finally become a polypropylene shopping bag (CO_2 valorisation plant on Figure 14). At the end-of-life, the shopping bag is incinerated with CCS to allow for the possible generation of negative emissions. All the inventory data are taken from the literature and provided in the excel file of the supplementary information (SI).

The inventory of miscanthus production is taken from the work of Jury et al. (2022). The soil organic carbon (SOC) changes due to miscanthus production is modelled with the AMG model (Clivot et al. 2019) over the entire lifespan of the plot. For the production of wood residues, the growth of trees is modelled using the Chapman-Richards equation and the parameters given Albers (2019) for the sessile oak. Sessile oak was chosen to obtain the most contrasting result possible compared to miscanthus. The frequency and amount of thinning are also taken from Albers (2019). Calculation details can be found in the SI 'LCI_from_excel_dyn.ipynb'. Emissions and consumptions linked to the production of biomass collected in the literature are representative of a production over the entire lifespan of a miscanthus or tree plot. The inventory of miscanthus production describes an almost constant production over 15 years. The inventory of wood residues describes a production every 5 to 10 years over 180 years, with a decreasing amount. For both miscanthus and wood residues, the temporal distribution of the biomass production is not equal to the temporal distribution of the biomass consumption in the step of fermentation. Thus, both the inventory of miscanthus and wood residues cannot be directly used as an input in the fermentation step. To overcome this issue, the dynamic LCI for the production of biomass over the entire lifespan of a plot is averaged to represent the mean production of one unit of biomass at $t_{0,process}$ following the algorithm illustrated in Figure 15.

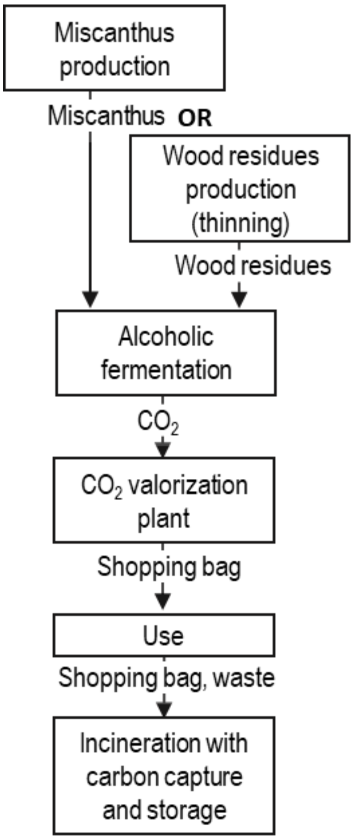

Figure 14: Life cycle steps of the case study

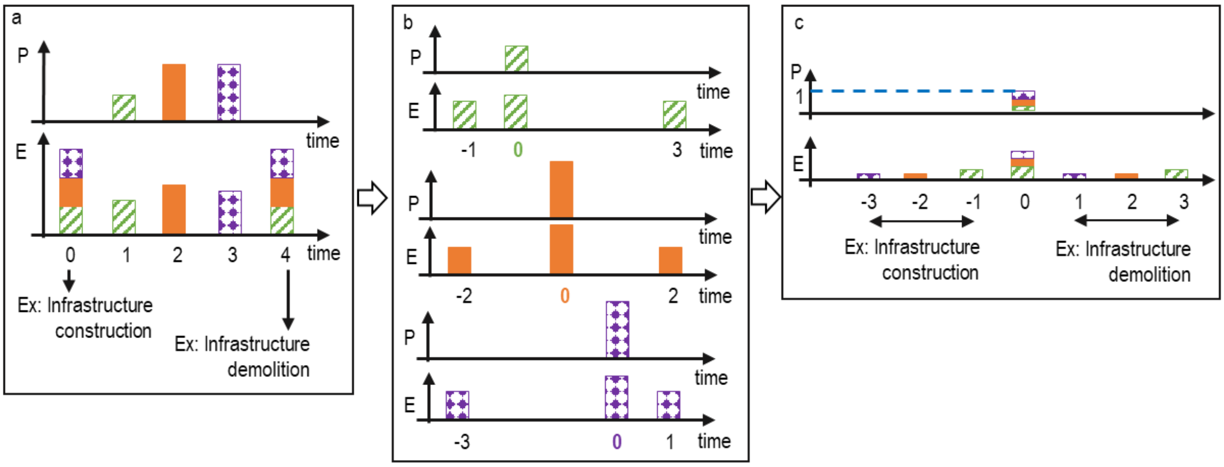

Figure 15: Illustration of the algorithm used for averaging a dynamic LCI. (a) Dynamic LCI representing the production over the entire lifespan of the system in chronological order. (b) The inventory is divided into one inventory by year of production. The pulse emissions/captures such as land use change or infrastructure construction are equally divided between the cycle of productions. The year of production becomes the $t_{0,\ process}$ of each new inventory. (c) The final averaged inventory representing the mean production at $t_{0,\ process}$ is then the average of the inventory by year of production weighted by the respective production volumes.

The cumulative radiative forcing induced by the system is calculated using the following formula:

$$AGWP(TH) = \sum_{i} \sum_{t_e} m_i(t_e) \int_{t_e+t_0}^{t_{end}} a_i C_i(t-t_e)dt \ = \sum_{i} \sum_{t_e} m_i(t_e) \int_{t_0}^{t_{end}-t_e} a_i C_i(t)dt \qquad (3)$$

With:

- i: greenhouse gas (CO_2, CH_4 or N_2O only)
- $t_{end} - t_0 = TH$: time horizon of the impact assessment (year). If not calendar based, $t_0 = 0$.
- t_e: time of emission or capture of a greenhouse gas (year). t_e can take values between $-\infty$ and t_{end}. When $t_e < t_0$, it implies that the emission or capture occurs before the time frame of the assessment. The integration time is then greater than TH. Beyond $t_{end,}$ the emissions or captures are cut-offed. They do not contribute to the radiative forcing.
- $m_i(t_e)$: mass of greenhouse gas i emitted at time t_e.
- a_i: radiative efficiency of the greenhouse gas i, based on AR5 values (IPCC 2013)($W.m^{-2}.kg^{-1}$).
- $C_i(t)$: decay function of the greenhouse gas i (yr^{-1}).

In the present article, dynamic modelling refers to the calculation of a dynamic LCI and its dynamic LCIA on climate change using Temporalis. Static modelling refers to using a static LCI, i.e. all emissions and consumptions occur at t_0, and performing LCIA on climate change for multiple time horizon using Temporalis.

3.2 MOTIVATION FOR THE CHANGES INTRODUCED IN TEMPORALIS

The calculation of the dynamic inventory from unit processes and the dynamic characterisation on climate change is performed using the version of Temporalis created by Cardellini et al. (2018).

A few changes have been made to the original source code. Firstly, as a graph traversal algorithm is used for calculating the dynamic LCI, and as an inventory in LCA can involve thousands of unit processes, a cut-off is applied to stop the graph traversal algorithm in order to limit the computing time. The balance between accuracy and computation time is well explained by Pigné et al. (2020). To enable the LCA practitioner to be aware of the magnitude of the impact not accounted for in the calculation, an attribute was added to the dynamic LCA object in Temporalis to store the cumulative impact of all the disregarded processes calculated with $AGWP_{100}$. Secondly, calculating the dynamic inventory can take up to several hours, depending on the complexity of the system and the performance of the computer. The code was modified to allow the storage of the calculated dynamic inventory into an excel file to be able to perform the characterisation of the inventory at a later date. Lastly, the code used to perform the characterisation of the dynamic inventory was simplified to make it easier for future users to understand and to limit integration errors. The analytical formula of the AGWP is directly used to calculate the radiative forcing induced by an emission rather than using numerical integration. The emission and capture of atmospheric CO_2 is characterised with the same function as fossil CO_2. The sign provided in the inventory indicates if it is an emission (plus) or a capture (minus). With this approach, there is also no need to differentiate atmospheric and fossil methane in the characterisation step. The modified source code is available in the SI 'Modified_version_temporalis.zip'.

3.3 SENSITIVITY ANALYSIS ON THE DEFINITION OF THE FUNCTIONAL UNIT

The production amount of the CO_2 valorisation plant is arbitrarily chosen, i.e. 1000 units per year for 20 years or 400 units per year for 50 years. As explained in the introduction, two functional units can be defined: "Production of 20000 bags over the entire lifespan of the plant (LP)" (FU_1) or "Production of 20000 bags at t_0" (FU_2). The dynamic LCI used to model FU_1 represents the entire system chronologically, for instance from infrastructure construction to infrastructure demolition for a production plant as illustrated in Figure 15a. The dynamic LCI used to model FU_2 is averaged as illustrated in Figure 15c. In static LCA, strictly the same results are obtained with both functional units. In a dynamic LCA approach, t_0 is defined as the time when the product, service or system is ready to be used as proposed by Beloin-Saint-Pierre et al. (2020). This definition leaves several possibilities to position the dynamic LCI relatively to t_0 in the case of the "Production of 20000 bags over LP". t_0 can correspond to any year between the first year of production (noted P_0) and the last year of production (noted P_{end}). To explore the impact of the definition of the functional unit and the position of the dynamic LCI relatively to t_0, results are calculated for the two functional units. Moreover, for the "Production of 20000 bags over LP" functional unit, the results are calculated for a lifespan of 20 or 50 years and for the two extreme temporal positions of the inventory, i.e. $P_0 = t_0$ or $P_{end} = t_0$. The lifespan of the plant does not change the results calculated with the "Production of 20000 bags at t_0" functional unit because the infrastructure construction and decommissioning are not included in the inventory due to lack of inventory data.

3.4 SENSITIVITY ANALYSIS: VARIATIONS INDUCED BY DYNAMIC MODELLING VS UNCERTAINTY IN STATIC ASSESSMENT

To limit the number of varying parameters, the functional unit used for performing this sensitivity analysis is chosen to be "Production of 1 bag at t_0". The only temporal parameter is thus the lifespan of the bag, either 0 or 20 years. A preliminary sensitivity analysis is performed in static LCA to select key parameters to the impact variation on climate change.

To identify the main contributors to the variation of static results, a parameterized model is built using lca-algebraic. Lca-algebraic (https://github.com/oie-mines-paristech/lca_algebraic) is a package of Brightway2 specific for uncertainty analysis. A few parameters are selected based on the variability observed during data collection and presented in Table 12, except for the range of values for the carbon content of biomass defined as ± 10% of the default value. Every distribution is assumed to be uniform due to lack of information. "Energy production" is a Boolean parameter. The first alternative corresponds to conventional energy production (EP_{conv}), with heat and hydrogen production modelled by Ecoinvent datasets (Ecoinvent) ("market for heat, from steam, in chemical industry" and "market for hydrogen, liquid"). For the second one, the amount of heat and hydrogen is set to zero to simulate a perfectly decarbonised production (EP_{zero}). Sobol indices are calculated to evaluate the contribution of each parameter uncertainty to the total model variance (Sobol 2001). First-order Sobol indices determine the individual contribution of parameters to the total model variance. Higher-order Sobol indices determine the contribution of interaction of multiple parameters to the total model variance. The sum of all Sobol indices is 1. The closer the Sobol index is to 1, the greater is the contribution of the parameter uncertainty to the total variance of the model.

Table 12: Parameters selected to perform a sensitivity analysis in static LCA. SOC: Soil Organic Carbon.

Parameter name	Default	Minimum	Maximum	Unit
Energy production	EP_{conv} or EP_{zero}			unitless
Carbon content miscanthus	0.48	0.43	0.52	$kg_C/kg_{biomass,\ dry\ matter}$
Carbon content wood residues	0.50	0.45	0.54	$kg_C/kg_{biomass,\ dry\ matter}$
SOC miscanthus	0.21	-0.09	0.5	$kg_{CO2}/kg_{miscanthus,\ dry\ matter}$
Stoichiometry fermentation	0.04	0.03	0.04	kg_{CO2} $/MJ_{ethanol}$
Yield fermentation	6	6	10	$MJ_{ethanol}/kg_{biomass,\ dry\ matter}$
CO_2 to methanol	1.45	1.37	1.84	$kg_{CO2}/kg_{methanol}$
Methanol to propylene	2.89	2.8	3.02	$kg_{methanol}/kg_{propylene}$
Heat efficiency CO_2 capture	3.7	2.95	7.52	$GJ/t_{CO2,\ captured}$
Yield CO_2 capture	0.9	0.9	1	$kg_{CO2,\ captured}/kg_{CO2,\ treated}$

4 RESULTS

For the sake of conciseness, the system producing a bag from CO_2 captured from miscanthus fermentation is referred to as “system producing a bag from miscanthus”. Similarly, the system producing a bag from CO_2 from the fermentation of wood residues is referred to as “system producing a bag from wood residues”.

In the section 4.1, resources created to facilitate the use of Temporalis are presented. In subsections 4.2 and 4.3, the results of the sensitivity analyses are presented for the production of bags from miscanthus, for their production from wood residues, and finally for the comparison of both systems (miscanthus minus wood residues).

4.1 RESOURCES TO FACILITATE THE USE OF TEMPORALIS

The modified version of Temporalis can be used to carry out a dynamic LCA, as illustrated by the results in the following subsections. All the documents created to carry out this dynamic LCA (jupyter notebooks, excel) are provided in SI. These documents can be used as inspiration to facilitate future dynamic LCA with Temporalis. The script for averaging a dynamic LCI (cf. Figure 15) is available in the SI 'LCI_from_excel_dyn.ipynb'. This SI also offers an example of the construction of unit processes containing temporal information. The SI 'Calculation_inventory.ipynb' shows how to calculate the dynamic inventory and store it in an excel file for future characterisation. The SI 'SA_dynVSstat.ipynb' contains examples of how to visualise contributions by groups of activities and by substances over time. It also contains an example to search for information in the calculated inventory.

4.2 SENSITIVITY ANALYSIS ON THE DEFINITION OF THE FUNCTIONAL UNIT

Beyond a TH value of 50 years, the results of the comparison using FU_1 (miscanthus minus wood residues) are within a narrow uncertainty range of ± 5% compared with the averaged approach (FU_2) (Figure 3). This comparison is dominated by the cumulative radiative forcing induced by the production from wood residues. In particular, the main contributor to its impact is the CO_2 captured by photosynthesis during tree growth. The temporal scope of such capture is at least three times longer (180 years) than the temporal scope of bag production (< 50 years), as illustrated in Figure 17. This reduces the influence of the definition of the functional unit on the results when looking at least after the first year of production. Figure 18 indicates that the dynamic cumulative radiative forcing induced by the production of bags from miscanthus (FU_1) is within an uncertainty range of ± 10% from the results obtained with the averaged approach (FU_2) only for TH superior to 100 years. Due to the shorter temporal scope of biomass production, this system is more dependent on the definition of the functional unit.

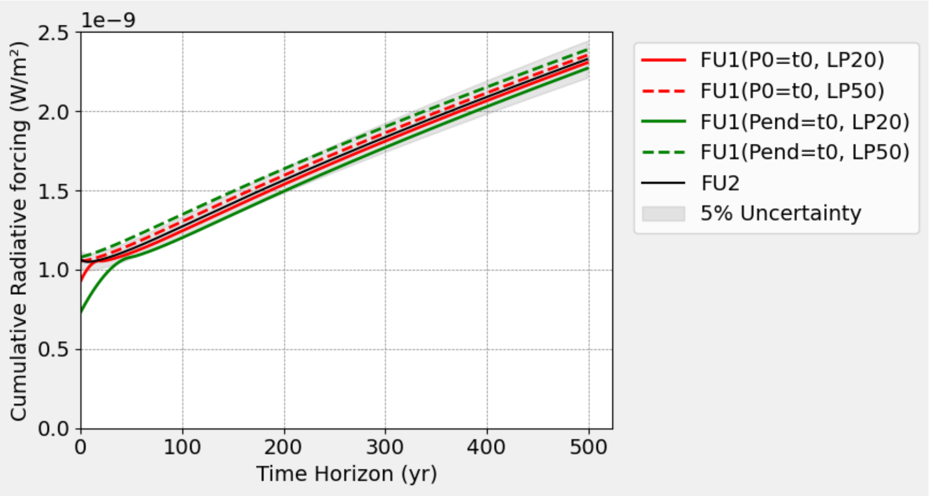

Figure 16: Evolution of the difference between the radiative forcing caused by the production of bags from miscanthus and the production of bags from wood residues (miscanthus minus wood residues). FU_1: "Production of 20000 bags over LP". The dynamic inventory is positioned relatively to t_0 either with the first year of production equal to t_0 ($P_0 = t_0$) or the last year of production equal to t_0 ($P_{end} = t_0$). LP: lifespan of the plant. FU_2: "Production of 20000 bags at t_0". The ± 5% of uncertainty is calculated on the results obtained with FU_2.

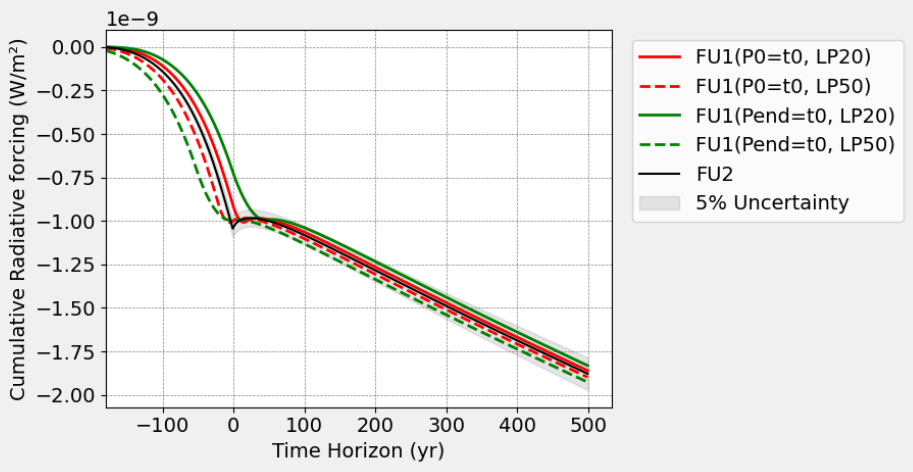

Figure 17: Evolution of the radiative forcing caused by the production of bags from wood residues. FU_1: "Production of 20000 bags over LP". The dynamic inventory is positioned relatively to t_0 either with the first year of production equal to t_0 ($P_0 = t_0$) or the last year of production equal to t_0 ($P_{end} = t_0$). LP: lifespan of the plant. FU_2: "Production of 20000 bags at t_0". The ± 5% of uncertainty is calculated on the results obtained with FU_2.

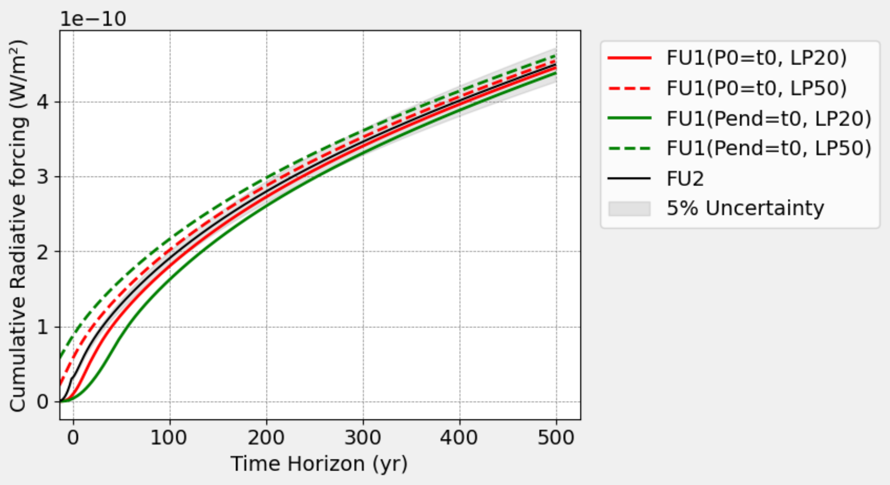

Figure 18: Evolution of the radiative forcing caused by the production of bags from miscanthus. FU_1: "Production of 20000 bags over LP". The dynamic inventory is positioned relatively to t_0 either with the first year of production equal to t_0 ($P_0 = t_0$) or the last year of production equal to t_0 ($P_{end} = t_0$). LP: lifespan of the plant. FU_2: "Production of 20000 bags at t_0". The ± 5% of uncertainty is calculated on the results obtained with FU_2.

Figure 16, Figure 17 and Figure 18 have in common the following aspects:

- For the same time horizon TH, by denoting I_{FU} the impact of the corresponding functional unit:

$$\lim_{TH \to +\infty} \frac{I_{FU1(P_0,TH)} + I_{FU1(P_{end},TH)}}{2} = \lim_{TH \to +\infty} I_{FU2(TH)} \tag{4}$$

when TH tends towards infinity, the impact calculated using FU$_2$ ("Production of 20000 bags at t_0") is equal to the average of the calculated impact using FU$_1$ ("Production of 20000 bags over LP") with $P_0 = t_0$ and $P_{end} = t_0$.

- By defining three different time horizons TH_1, TH_2 and TH_3, then

$$I_{FU1(P_0,TH_1)} = I_{FU1(P_{end},TH_3)} = I_{FU2(TH_2)} \xRightarrow[TH\rightarrow +\infty]{} TH_2 = TH_1 - \frac{LP}{2} = TH_3 + \frac{LP}{2} \qquad (5)$$

when TH tends to infinity, the impact calculated using FU$_2$ ("Production of 20000 bags at t_0") is equivalent to the impact calculated using FU$_1$ ("Production of 20000 bags over LP") with t_0 positioned at the middle of the production time ($\frac{LP}{2}$).

This results from the fact that CO$_2$ emissions are the main contributor to the total impact, and from two modelling choices made. Firstly, the production of 1 unit of product is modelled with the same temporal distribution of emissions for both types of functional unit. Secondly, the total production of 20000 units is uniformly distributed over the lifespan of the plant. The graphical observations are mathematically verified using a simple system emitting a total mass of CO$_2$ uniformly over the lifespan of the system, see SI named 'SI_1.docx' (Annexe 2).

4.3 SENSITIVITY ANALYSIS: VARIATIONS INDUCED BY DYNAMIC MODELLING VS UNCERTAINTY IN STATIC

In the subsection 4.3.1, the results of the sensitivity analysis on static results are presented to select key parameters influencing the calculation of the climate change impact. In the subsection 4.3.2, the results of the sensitivity analysis between dynamic and static modelling are presented.

4.3.1 Selection of key parameters contributing to the static impact variation on climate change

The first-order Sobol indices of each parameter are summarised in Table 13. More than 94% of the variance is explained with first-order Sobol indices, so higher-order Sobol indices are not calculated. Table 13 reveals that the variation of 'energy production' explains most of the results variation for the system producing a bag from wood residues and half of the results variation for the system producing a bag from miscanthus. The other half is explained by the variation of SOC change.

Table 13: First-order Sobol indices for each parameter selected to perform an uncertainty analysis. SOC: Soil Organic Carbon.

Parameter name	Miscanthus	Wood residues
Energy production	0.48	0.94
Carbon content miscanthus	0.01	-
Carbon content wood residues	-	0
SOC miscanthus	0.46	-
Stoichiometry fermentation	0	0
Yield fermentation	0.01	0
CO2 to methanol	0.01	0
Methanol to propylene	0	0
Heat efficiency CO_2 capture	0.02	0.02
Yield CO_2 capture	0	0
Sum of the first-order Sobol indices	0.99	0.94

To summary, in the following subsection, to compare the variation induced by dynamic LCIA and the variations induced by uncertainty on inventory data, the impact on climate change is calculated for every combination of the parameters values:

- LCI modelling and impact characterisation: static or dynamic,
- LB (lifespan of the bag): 0 or 20 years,
- EP (Energy production): EP_{conv} or EP_{zero},
- SOC changes: high or low.

4.3.2 Results sensitivity analysis dynamic vs static

The results for wood residues are shown in Figure 19 and for miscanthus in Figure 20. Figure 21 shows the comparison between the two biomass sources.

Two sets of curves stand out on the Figure 19. The first set have an impact of 0 W/m² for TH equal to zero. It regroups the results calculated with static LCI. The second set have an impact of around 5×10^{-14} W/m² for TH equal to zero. It regroups the results calculated based on a dynamic LCI. At a TH of 100 years, the variation due to the choice between static and dynamic modelling is around 4×10^{-14} W/m². At $TH = 100\ years$, the variation due to the uncertainty on static inventory data (EP_{conv} vs EP_{zero}) is of the same order of magnitude, around 5×10^{-14} W/m². The variation due to the uncertainty on static inventory data is increasing over time due to the cumulative nature of the AGWP. However, the variation due to the choice between static and dynamic modelling remains relatively stable over time. Due to the fact that CO_2 is the main contributor to the impact, when TH tends towards infinity, the difference between static and dynamic modelling tends to $a_{CO2}a_0\sum_{te}m_e t_e$, with a_{CO2} the radiative efficiency of CO_2, a_0 the first coefficient of the decay function of CO_2, and m_e the mass of CO_2 emitted at time t_e (demonstration included in the SI named 'SI_1.docx'). Using a simplified emission pattern (uniform CO_2 capture over 180 years) the calculated difference between static and dynamic modelling for the system using wood residues is 3×10^{-14} W/m². This is of the same order of magnitude than the asymptotic difference observed in Figure 19 when TH tends towards infinity (static (EP_{conv}) minus dynamic (EP_{conv}) or static (EP_{zero}) minus dynamic (EP_{zero})). The difference between static and dynamic modelling due to the lifespan of the bag when TH tends towards infinity is negligible, at approximately 5×10^{-16} W/m².

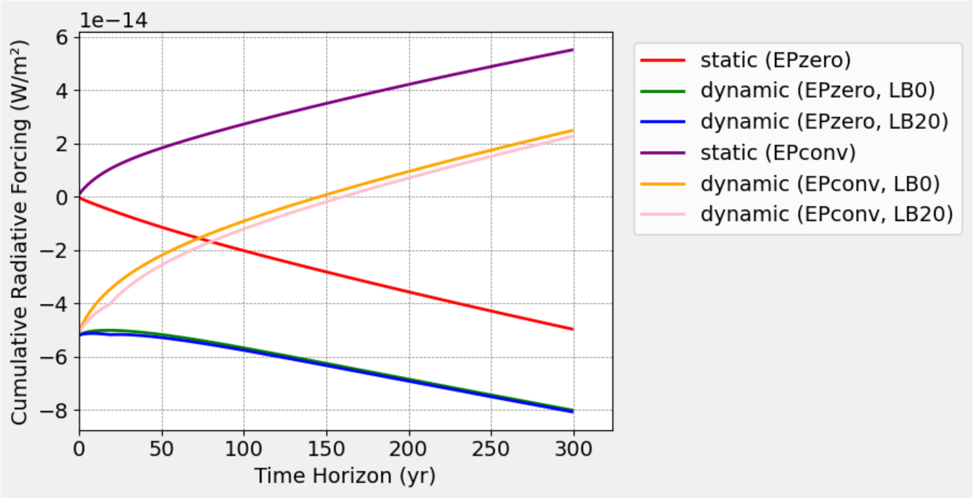

Figure 19: Evolution of the radiative forcing caused by the production at t_0 of one bag from wood residues. EP: Energy production, LB: Lifespan of the bag.

Four sets of curves stand out on Figure 20. They are directly related to the values of the static parameters: energy production (EP) and SOC changes. The curves calculated with static and dynamic LCIs overlap. The temporal distribution of the mean SOC changes for miscanthus production is symmetrical around t_0. Subsequently, the term $\sum_{te}m_e t_e$ related to SOC changes is equal to zero. When TH tends towards infinity, there is no variation due to the choice between static and dynamic modelling.

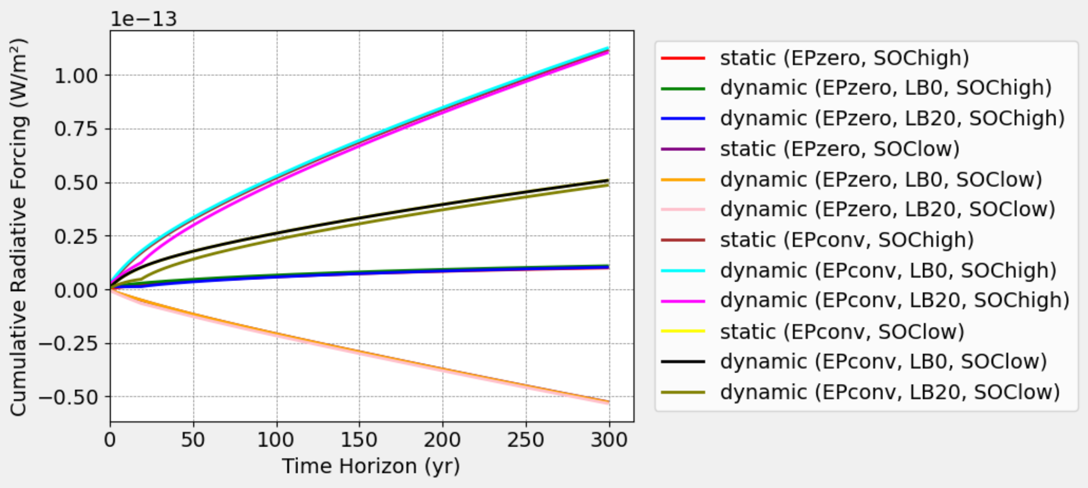

Figure 20: Evolution of the radiative forcing caused by the production of one bag at t_0 from miscanthus. EP: Energy production, LB: Lifespan of the bag, SOChigh: Scenario where miscanthus production leads to soil organic carbon stock decrease. SOClow: Scenario where miscanthus production leads to soil organic carbon stock increase.

The parameter LB, lifespan of the bag, have no influence on the results illustrated in Figure 21. The parameter LB is linked to the bag's end-of-life which is identical in both systems, causing the same impact variation. The energy production parameter EP is used in the calculation of the LCI of several identical life cycle steps between the compared system (CO_2 transformation into a bag, CO_2 capture after the bag incineration), and also in the LCI of the fermentation step. The carbon content of wood residues is different from that of miscanthus, leading to a different yield of CO_2 production during the fermentation step. This explains the small variation due to the parameter EP when comparing the two systems. The variation due to dynamic modelling is strongly dominated by the impact variations of the wood residues system, as illustrated in Figure 19 and Figure 20.

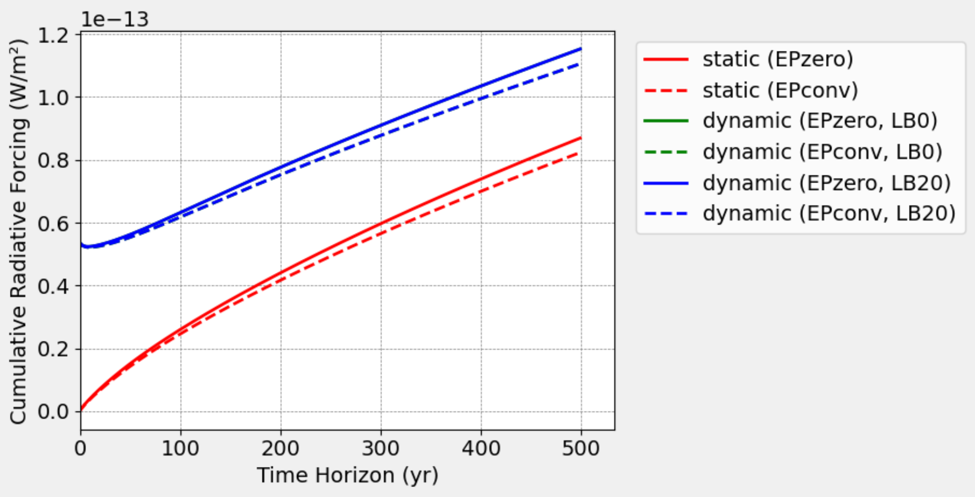

Figure 21: Evolution of the difference between the radiative forcing caused by the production of bags at t_0 from miscanthus and the production of bags at t_0 from wood residues. EP: Energy production, LB: Lifespan of the bag. The miscanthus production leads to a decrease in SOC stock (SOChigh).

5 DISCUSSION

In section 5.1, the usefulness of Temporalis and prospects for improvement are discussed. In section 5.2, the results of the sensitivity analysis on the definition of the functional unit are exploited to propose recommendations for harmonizing their definition, facilitating future interpretation and comparison of dynamic LCIA results and studies. In section 5.3, sensitivity analysis on dynamic modelling versus uncertainty on static parameters are discussed with regards to the method proposed by Collet et al. (2014) for selecting flows for which it is important to add temporal information.

5.1 TEMPORALIS – FEEDBACK AND OUTLOOK

It should be noted that there is a promising ongoing project to update Temporalis (https://github.com/brightway-lca/bw_temporalis). In the meantime, the modified version of Temporalis provided in the SI of this article is a working tool for dynamic LCA. Nevertheless, there is still room for improvement. Firstly, the modified script could be improved further by offering the possibility to account for the cut-offed unit processes in a static way. Such approach assumes that the emissions due to the entire life cycle of the process (calculated with the usual matrix calculation) are emitted the same year as the year of consumption of the process. This would reduce the calculation error due to the stopping condition of the graph traversal algorithm. Secondly, only AGWP and AGTP using AR5 parameters for CO_2, CH_4 and N_2O without climate-carbon feedback are currently included as characterisation methods in the modified version of Temporalis. Including more characterisation formulas would be relevant to perform sensitivity analysis on the chosen metric. In fact, the background concentration of CO_2, CH_4, N_2O is steadily rising. The background concentration of CO_2 reached 410 ppm in 2019 leading to an update of the radiative efficiency of CO_2 in the latest IPCC report, but not of the decay function (IPCC 2021). Reisinger et al. (2011) and Caldeira and Kasting (1993) demonstrated that an increase in the CO_2 background concentration led to a decrease in the radiative efficiency and an increase in climate-carbon cycle feedback, both effects partially cancelling each other out. The decay function should thus also be updated to not underestimate the impact of an emission of CO_2 on climate change.

In dynamic LCA, implementing characterisation factors that depend on the evolution of the background concentration of CO_2 would imply to use a different AGWP formula for each time of emissions. The AGWP formula would depend on the initial background concentration of CO_2 and its subsequent prospective evolution. This seems too complex relative to the gain in precision. A more general examination of how to account for the uncertainty of the characterisation factors in LCA seems more useful to address this issue. Thus, in Temporalis, AGWP and AGTP could be proposed with or without climate-carbon feedback, and using AR5 or AR6 parameters to be able to perform a sensitivity analysis on the metric used. Lastly, some indicators can be deduced from metrics such as AGTP. AGTP can be used for calculating indicators such as the amplitude of the temperature change or years of temperature peaks as developed for instance by Tiruta-Barna (2021). Script could also be written to calculate such indicators from the characterised inventory.

5.2 SENSITIVITY ANALYSIS ON THE DEFINITION OF THE FUNCTIONAL UNIT

The results obtained from the case study are used to formulate more general recommendations. The particularity of the case study is that the same dynamic LCI is used for modelling the production of 1 unit for both functional unit (FU_1 and FU_2). It corresponds to a dynamic LCI that does not include pulse emissions, such as large infrastructure construction or land use change (cf. the algorithm to create an average dynamic LCI, see Figure 15). For systems sharing this particularity, the comparison results obtained with the two functional units are almost equivalent (less than 5% of the difference for TH superior to the lifespan of the plant), as observed in section 4.2.

The following considerations are applicable to all types of systems. The potential impact on climate change of a given entire system is evaluated by using the following functional unit: “production of several units of the product or service each year over the entire lifespan of the system”. Such functional unit is relevant to evaluate a system relative to specific climate goals. Climate goals are defined for calendar-based time horizons, which resolves the ambiguity identified in the position of the dynamic LCI relative to t_0. For instance, climate neutrality needs to be reached by 2050 to limit global warming at 1.5°C (IPCC 2018).

However, the position of the dynamic LCI relative to t_0 ($P_0 = t_0$ and $P_{end} = t_0$) might have an influence when comparing to static results. This depends on the distribution of emissions contributing to the impact. If the majority of emissions occurs periodically over the lifespan of the plant (LP), LP is the longest temporal scope included in the LCI. The longer it is, the greater the difference in results depending on the position of the dynamic LCI relative to t_0 ($P_0 = t_0$ and $P_{end} = t_0$). This is illustrated by the case study with miscanthus (Figure 18). For a $LP = 20\ years$, the results are within the ± 5% window after a time horizon of around 100 years. With $LP = 50\ years$, this period increases to around 250 years. However, if LP is not the longest temporal scope, its influence is reduced. This is illustrated by the case study with wood residues: the CO_2 is captured over a much longer temporal scope than LP (180 years as opposed to 20 years or 50 years). The results are within the ± 5% window after a time horizon equal to LP, see Figure 17. In conclusion, if the time horizon is much longer than LP, the chosen position of the dynamic LCI relative to t_0 ($P_0 = t_0$ and $P_{end} = t_0$) will not influence the comparison to static results. If the time horizon is not much longer than LP and LP is the longest temporal scope included in the LCI, then the position of the dynamic LCI relative to t_0 will influence the comparison to static results and should be clearly stated when communicating the results. $P_{end} = t_0$ is more coherent with the static interpretation of the time horizon. In static LCA, the results represent the potential impact at a given time horizon of delivering the functional unit. The functional unit is entirely delivered only after the last year of production in dynamic LCA.

The "production of several units of the product or service at t_0" functional unit is relevant to compare systems that do not share the same temporal distribution of production. For example, as explained in section 3.1, the production of miscanthus do not share the same temporal distribution of production as the production of wood residues but an LCA practitioner might want to compare the impact of producing 1 kg of miscanthus with the impact of producing 1 kg of wood residues. Moreover, the inventory data can be reused as background inventory data in another life cycle.

5.3 SENSITIVITY ANALYSIS: VARIATIONS INDUCED BY DYNAMIC MODELLING VS UNCERTAINTY IN STATIC

The results indicate that the variation induced by dynamic modelling is significant for wood residues production compared to the variations induced by uncertainty on energy production modelling. However, the variation induced by dynamic modelling is not significant for miscanthus production compared with the variations induced by uncertainty on energy production and SOC changes. Based on the method of Collet et al. (2014), the variation of SOC stock was relevant for two reasons. Firstly, the results are sensitive to a variation of the initial value of SOC stock as demonstrated in Figure 20. Secondly, the variations of SOC stock during miscanthus production are distributed over 30 years, more than the temporal resolution of climate change identified as one year. Collet et al. (2014) proposed a method applicable to every impact category. Examining the mathematical formula of each characterisation factor in depth was out of the scope of their study. As demonstrated in section 3.2, information on the magnitude of variations induced by dynamic modelling can be calculated using simplified formulas constructed from the study of the AGWP when TH tends towards infinity. Further study of the mathematical properties of AGWP seems like a promising idea to improve the method with a focus on climate change. If the goal of the dynamic LCA is to compare systems, it is unnecessary to add temporal information to identical steps for both systems. This will not change the conclusion of the comparison.

6 CONCLUSION

Temporalis is an efficient tool to perform dynamic LCIA. Two areas for improvement were identified: dealing with the loss of information due to the cut-off included in the graph-traversal algorithm and proposing more characterisation methods to be able to perform a sensitivity analysis.

The "production of several units of the product or service each year over the entire lifespan of the system" functional unit should be used for evaluating the potential impact on climate change of the entire system relative to climate goals on a calendar-based timeline. To compare the obtain results to static results, the TH should be defined starting from the last year of production. The temporally averaged functional unit ('1 unit produced at t_0') should be used for comparing systems that do not share the same temporal distribution of production and to build inventory data reusable as background inventory data in another life cycle.

Being able to pinpoint which flow will benefit from being distributed on a timescale is important for a LCA practitioner to save time for improving a static inventory and perform sensitivity analysis. Further research on the mathematical properties of AGWP would enable to improve the method proposed by Collet et al. (2014) to construct a method for choosing which flow to distribute on a timescale prior to a full dynamic LCIA using only simplified temporal information on a given system.

CHAPTER CONCLUSION

This chapter provides recommendations, on the functional units as well as clarification on how to position the inventory timeline relative to the impact characterisation timeline, that can be applied to the systems described in chapter 1.

Firstly, including time in the assessment allows to further explore the question "Is the studied system environmentally beneficial being compared to business as usual?" (cf. chapter 2). On a calendar-based timeline, using the "total amount of units produced over the entire lifespan of the system" functional unit, three options can be compared: i) the conventional system with an extension of its lifespan through maintenance (business as usual only), ii) the conventional system until its planned end-of-life then replaced by the CCUNET system and iii) the immediate replacement of the existing plant by the CCUNET system. Concerning the second question "is it environmentally better to capture, use and then permanently store the CO_2 (CCUNET), or to capture and permanently store the CO_2 (CCS)?", it is answered by comparing two future systems sharing the same production temporal distribution, so the "total amount of units produced over the entire lifespan of the system" functional unit can also be used.

Secondly, the proposed algorithm for averaging a dynamic LCI is compatible with the evaluation of negative emissions. Allocation is performed to distribute emissions between years of productions but it does not lead to exclusion of emissions or captures. The results revealed that if the biomass take decades to growth, the potential for negative emissions is underestimated using static LCA. The AGWP is used to assess marginal changes in relation to a background situation assumed to be constant, as usually done in LCA. To perform a meaningful evaluation relative to a defined climate goal (e.g. climate neutrality by 2050), the system boundaries of the given system should only include future emissions or captures. Future emissions or captures occur after the reference year used for calculating the characterisation factors and climate goal. It corresponds to 2019 when using the parameters provided in the latest report of the IPCC .

Lastly, an additional goal is also to compare NETs systems to support decision-making by allowing meaningful benchmarking, see section 4.1 in chapter 2. The functional unit “kg of CO_2 treated” can have very different temporal distribution depending on the NET system. For example, the temporal scope of an afforestation project is of several decades, depending on the species chosen, until the forest reaches a saturation point. The amount of CO_2 treated each year depends on the growth rate of trees. The temporal scope of a BECCS project is equal to the lifespan of the bioenergy plant. The amount of CO_2 treated each year depends on the amount of energy produced. Thus, to perform a relevant comparison of NET systems, the functional unit “1 kg of CO_2 treated at t_0” should be used as it will include the timescale discrepancies in the assessment through the averaged LCI.

The operational version of Temporalis, the algorithm to average dynamic LCI and the reusable example provided in this chapter will hopefully facilitate the use of dynamic LCA to perform such comparison. However, the results also show that the effort required to carry out dynamic LCA does not necessarily lead to very different results from static LCA. To enable LCA practitioners to target their efforts more effectively, a method is necessary for deciding if the assessment requires further investigation using dynamic LCA. Such a method is proposed in the following chapter.

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Chapter 4: Is dynamic LCA necessary? evaluation with simplified temporal information

A LCA practitioner has limited time and might need to choose between improving his inventory data in static LCA or perform dynamic LCA. The previous chapter illustrates that dynamic LCA demands more data on the system and increases the complexity of the calculation, but the precision of the results is not necessarily significantly increased. Hence the question raised in this chapter: with a focus on climate change, is it possible to determine if the dynamic approach will significantly change the results compared to a static approach with only a little knowledge of the dynamics of the system under study? To answer this question, the problem is first expressed mathematically in section 1. Then, in section 2, the mathematical expressions are calculated for AGWP. In section 3, the method is applied to some examples. In section 4, threshold values of pattern duration above which dynamic LCA is required are provided for a limited number of emissions patterns.

List of abbreviations

(A)GWP	(Absolute) Global Warming Potential
CF	Characterisation Factors
LCA	Life Cycle Assessment
TH	Time Horizon

1 MATHEMATICAL EXPRESSION OF THE PROBLEM

Results calculated using a dynamic approach (I_{dyn}) can be compared with results obtained using a static approach (I_{stat}) by looking at the ratio between these results, i.e. I_{dyn}/I_{stat}. The closer the ratio is to one, the lesser the dynamic approach is relevant. However, the goal of an LCA is not only to calculate a final score, but also to know the main contributors. For example, if the ratio is close to 1, but as a result of a trade-off between a significant increase in the impact of CO_2 and a significant decrease in the impact of CH_4, it is worthwhile to further investigate using dynamic LCA. Indeed, CO_2 and CH_4 have very different lifetime in the atmosphere, so dynamic LCA can be used to reveal temporal trade-offs (cf. section 4.3 of the general introduction). Therefore, to determine whether it is necessary to further investigate using dynamic LCA, it is more relevant to calculate I_{dyn}/I_{stat} substance by substance.

Dynamic systems are essentially continuous, but for the sake of simplicity, the reasoning is presented using a discrete representation of time. An annual time step is chosen, corresponding to climate dynamics. The temporal differentiation[2] of each GHG emissions is thus modelled by discrete emissions patterns, as done in Temporalis. The ratio for a given GHG_i is then mathematically expressed as follows:

$$\left[\frac{I_{dyn}}{I_{stat}}\right]_i = \frac{\sum_{t_e}[m_i(t_e) \times CF_i(TH - t_e)]}{m_{i,total} \times CF_i(TH)} \tag{1}$$

which can be written as

$$\left[\frac{I_{dyn}}{I_{stat}}\right]_i = \sum_{t_e}\left[\frac{m_i(t_e)}{m_{i,total}} \times \frac{CF_i(TH - t_e)}{CF_i(TH)}\right] \tag{2}$$

With:

- $m_i(t_e)/m_{i,total}$: mass ratio between the amount of GHG_i emitted at time t_e and its total amount emitted over the whole life cycle. This represents the relative emissions over time. Simple emissions patterns are proposed in the following subsection to enable the calculation of this ratio using as little temporal information as possible.
- $CF_i(TH - t_e)/CF_i(TH)$: *r*atio of the dynamic characterisation factor for an emission at t_e on the static characterisation factor for the same time horizon TH. This corresponds to the variation in the characterisation factor due to the inclusion of temporality. This ratio is calculated numerically, using a python script.

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[2] "The action of distributing the information on a time scale related to the models' components. For example, elementary flows could be described per day or year. Different processes representing yearly average are another example" Beloin-Saint-Pierre et al. (2020).

In the following section, a given emission pattern is a succession of "pulse" emissions of a single type of GHG, the subscript i is thus omitted for clarity reasons.

1.1 EMISSIONS PATTERNS FOR THE CALCULATION OF THE MASS RATIO USING AS LITTLE INFORMATION AS POSSIBLE

The goal of this method is not to obtain a precise result but to assess whether it is worth to further investigate using dynamic LCA. The decision must be based on temporal information that is easy to collect. Two types of emissions pattern are thus translated mathematically to describe systems: a "pulse" emission (i.e. a peak emission at a time t), and a "linear" emissions pattern. A "linear" emissions pattern is a succession of "pulse" emission emitted each year $t_e \in [t_{min}, t_{max}]$, which intensity $m_p(t_e)$ for the pattern p varies monotonously between $m_p(t_{min})$ and $m_p(t_{max})$. The "uniform" emissions pattern (succession of "pulse" emissions of equal intensity) is a specific case of the linear emissions pattern, i.e. $m_p(t_{min}) = m_p(t_{max})$. As the emissions pattern is discretized on a yearly basis, $n_{pulses} = t_{max} - t_{min} + 1$ represents the actual number of pulse emissions included in the "linear" emissions pattern and is a dimensionless value. The total mass of GHG$_i$ emitted for a linear emissions pattern is:

$$m_{p,total} = n_{pulses} \times \left(\frac{m_p(t_{max}) + m_p(t_{min})}{2} \right) \tag{3}$$

The mass emitted at t$_e$ can be expressed as follow

$$m_p(t_e) = \frac{m_p(t_{max}) - m_p(t_{min})}{t_{max} - t_{min}} (t_e - t_{min}) + m_p(t_{min}) \tag{4}$$

It should be noted that if $m_p(t_{min})$ (or $m_p(t_{max})$) is equal to zero, the mass ratio $m_p(t_e)/m_{p,total}$ is independent of the value of $m_p(t_{max})$ (or $m_p(t_{min})$). If $m_p(t_{min}) = 0$:

$$\frac{m_p(t_e)}{m_{p,total}} = \frac{\frac{m_p(t_{max}) - 0}{t_{max} - t_{min}} (t_e - t_{min}) + 0}{n_{pulses} \times \left(\frac{m_p(t_{max}) + 0}{2} \right)} = 2 \times \frac{t_e - t_{min}}{(t_{max} - t_{min}) \times n_{pulses}} \tag{5.1}$$

If $m_p(t_{max}) = 0$:

$$\frac{m_p(t_e)}{m_{p,total}} = \frac{\frac{0 - m_p(t_{min})}{t_{max} - t_{min}} (t_e - t_{min}) + m_p(t_{min})}{n_{pulses} \times \left(\frac{0 + m_p(t_{min})}{2} \right)} = 2 \times \frac{t_{max} - t_e}{(t_{max} - t_{min}) \times n_{pulses}} \tag{5.2}$$

An entire lifecycle consists of a succession of activities. Each activity can generate a different emissions pattern for the studied GHG$_i$. For instance, removal of atmospheric CO$_2$ during biomass growth, the latter harvested at t_0, will be modelled by a "linear" emissions pattern while the biomass harvesting will be modelled by a "pulse" emission of CO$_2$ at t_0. Let

$$m_{total} = \sum_p m_{p,total} = \sum_p \sum_{t_e} m_p(t_e) \tag{6}$$

Equation (2) can thus be written as:

$$\left[\frac{I_{dyn}}{I_{stat}}\right] = \sum_{t_e}\left[\sum_p \frac{m_{p,total}}{m_{total}} \times \frac{m_p(t_e)}{m_{p,total}}\right] \times \frac{CF(TH - t_e)}{CF(TH)} \tag{7.1}$$

t_e and p are independent and can thus be reversed. Moreover, the $m_p(t_e)/m_{p,total}$ ratio is independent of t$_e$ and can be extracted from the sum on t_e, giving:

$$\left[\frac{I_{dyn}}{I_{stat}}\right] = \sum_p \frac{m_{p,total}}{m_{total}} \times \left[\frac{I_{dyn}}{I_{stat}}\right]_p \tag{7.2}$$

With $\left[I_{dyn}/I_{stat}\right]_p$ the ratio calculated for a given emissions pattern.

It should be noted that $\left[I_{dyn}/I_{stat}\right]_p \approx 1$ if the two following conditions are met:

- the emissions pattern is symmetrical around zero, i.e. $m_p(-t_e) = m_p(t_e)$, and all emissions are happening before TH and thus included in the impact assessment;
- the ratio of the characterisation factors can be approximated by a linear function of t$_e$ around the time of emission equal to zero, i.e. $CF_i(TH - t_e)/CF_i(TH) \approx at_e + b$ with $a \in R \text{ and } b = CF_i(TH - 0)/CF_i(TH) = 1$.

To explain this, the sum in equation (2) is split between the emissions occurring before $t_e = 0$, the emission at $t_e = 0$ and the emissions occurring after $t_e = 0$:

$$\left[\frac{I_{dyn}}{I_{stat}}\right]_p \approx \frac{1}{m_{p,total}}\left[\sum_{t_e=t_{min}}^{-1}\left[m_p(t_e)(at_e+1)\right] + m_p(0)(a \times 0 + 1) + \sum_{t_e=1}^{t_{max}}\left[m_p(t_e)(at_e+1)\right]\right] \tag{8.1}$$

We have

$$\sum_{t_e=t_{min}}^{-1}\left[m_p(t_e)(at_e+1)\right] = a\sum_{t_e=t_{min}}^{-1}\left[m_p(t_e)t_e\right] + \sum_{t_e=t_{min}}^{-1}\left[m_p(t_e)\right] \tag{9.1}$$

As $t_{min} = -t_{max}$ then:

$$\sum_{t_e=t_{min}}^{-1}\left[m_p(t_e)(at_e+1)\right] = a\sum_{t_e=-t_{max}}^{-1}\left[m_p(t_e)t_e\right] + \sum_{t_e=t_{min}}^{-1}\left[m_p(t_e)\right] \tag{9.2}$$

As $m_p(-t_e) = m_p(t_e)$ then:

$$\sum_{t_e=t_{min}}^{-1}\left[m_p(t_e)(at_e+1)\right] = -a\sum_{t_e=1}^{t_{max}}\left[m_p(t_e)t_e\right] + \sum_{t_e=t_{min}}^{-1}\left[m_p(t_e)\right] \tag{9.3}$$

And thus equation (8.1) becomes:

$$\left[\frac{I_{dyn}}{I_{stat}}\right]_p \approx \frac{1}{m_{p,total}}\left[-a\sum_{t_e=1}^{t_{max}}\left[m_p(t_e)t_e\right] + \sum_{t_e=t_{min}}^{-1}\left[m_p(t_e)\right] + m_p(0) + a\sum_{t_e=1}^{t_{max}}\left[m_p(t_e)t_e\right] + \sum_{t_e=1}^{t_{max}}\left[m_p(t_e)\right]\right] \tag{8.2}$$

$$\left[\frac{I_{dyn}}{I_{stat}}\right]_p \approx \frac{1}{m_{p,total}}\left[\sum_{t_e=t_{min}}^{-1}\left[m_p(t_e)\right] + m_p(0) + \sum_{t_e=1}^{t_{max}}\left[m_p(t_e)\right]\right] \approx \frac{1}{m_{p,total}}\sum_{t_e=t_{min}}^{t_{max}}\left[m_p(t_e)\right] \tag{8.3}$$

$$\left[\frac{I_{dyn}}{I_{stat}}\right]_p \approx 1 \tag{8.4}$$

This subsection provides emissions patterns for which the mass ratio depends only on one (“pulse” emission, symmetrical emissions pattern) to four temporal parameters (“linear” emissions pattern). This limits the data collection effort compared to a full dynamic LCA. In the following section, the ratio $CF_i(TH - t_e)/CF_i(TH)$ is calculated for the AGWP.

2 APPLICATION TO THE AGWP

The evolution of the ratio $CF_i(TH - t_e)/CF_i(TH)$ as a function of t_e (expressed as a fraction of TH) for CO_2 and CH_4, and for several time horizons is shown in Figure 22. Since CO_2 has an almost infinite lifetime and AGWP is a cumulative indicator, the impact of an emission of CO_2 accumulates indefinitely. This explains the shape of the curve: if CO_2 is emitted before t_0, it accumulates more impact than calculated with static LCA, and vice versa. On the other hand, CH_4 has a short lifetime in the atmosphere. CH_4 is thus less sensitive to dynamic modelling. To reach a dynamic characterisation factor more than 10% lower than the static characterisation factor, a peak emission of CH_4 needs to be emitted at least at 60% of the TH. In comparison, to reach the same results with a peak emission of CO_2, the time of emission only needs to be at 10% of the TH. It should be noted that CH_4 oxidises to CO_2. This explains the higher dynamic characterisation factor compared to static characterisation factor when CH_4 is emitted before t_0.

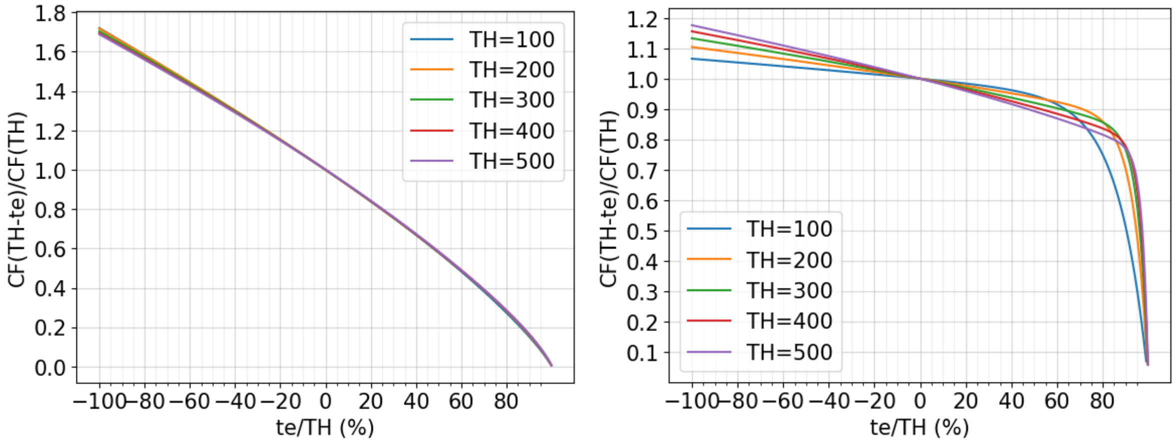

Figure 22: Evolution of the ratio $CF_i\,(TH\text{-}t_e)/CF_i\,(TH)$ as a function of t_e (expressed as a fraction of TH) for CO_2 (left) and CH_4 (right)

In Figure 22, the curves appear to be linear for $t_e/TH \in [-50\%, 50\%]$. If verified, a symmetrical emissions pattern around $te = 0$ with an half duration inferior to 50% of the TH would have a $[I_{dyn}/I_{stat}]$ ratio of approximately 1, without needing any further information for the calculation. To verify this information, a linear regression is performed on the $AGWP_i(TH - t_e)/AGWP_i(TH)$ curves for $TH = 100\ and\ 500$ and $i = CO_2\ and\ CH_4$. As illustrated in Figure 23, the quality of the linear approximation depends on the chosen interval for te. The $AGWP_{CO2}(100 - t_e)/AGWP_{CO2}(100)$ curve can be approximated by a linear function for $t_e/TH \in [-45\%, 45\%]$, see Figure 23 (R² = 0.999, b = 0.99). The linear approximation is of lower quality for $t_e/TH \in [-100\%, 100\%]$, notably for the value of b, as illustrated in Figure 23 (R² = 0.993, b = 0.96). The intervals for which the R² is greater than 0.999 and b included between 0.99 and 1 for the other curves are as follows:

- For $i = CO_2\ and\ TH = 500,\ t_e/TH \in [-40\%, 40\%]$
- For $i = CH_4\ and\ TH = 100,\ t_e/TH \in [-20\%, 20\%]$
- For $i = CH_4\ and\ TH = 500,\ t_e/TH \in [-40\%, 40\%]$

Except for CH_4 at a TH of 100 years, the R² is greater than 0.999 and b included between 0.99 and 1 for $t_e/TH \in [-40\%, 40\%]$. Extending the interval up to $t_e \in [-40\%, 40\%]$ for CH_4 at a TH of 100 years leads to R² = 0.994, b = 1. So if i) the GHG is either CO_2 or CH_4, ii) $TH \in [100, 500]$ and iii) the emissions pattern is symmetrical around zero with a half duration of less than 40% of the TH, it is not worth to further investigate using dynamic LCA.

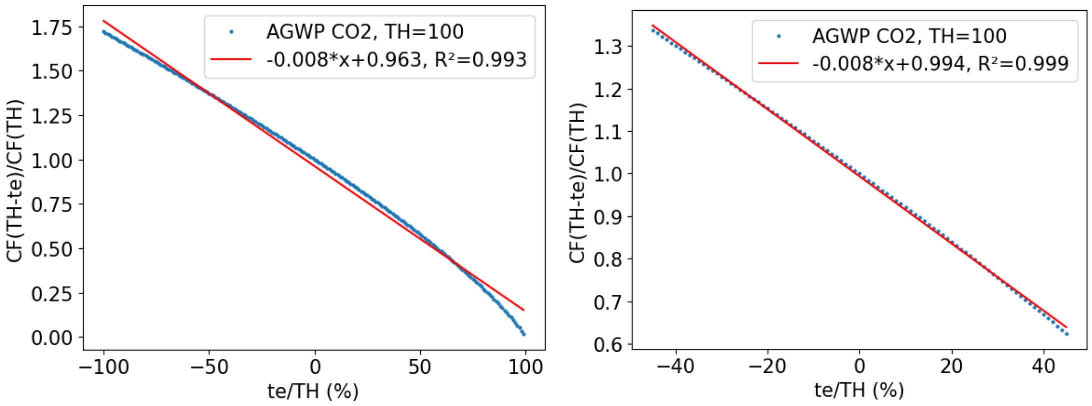

Figure 23: Evolution of the ratio AGWP$_{CO2}$ (100-t$_e$)/AGWP$_{CO2}$ (100) as a function of t$_e$ (expressed as a fraction of TH) for CO$_2$. The linear function obtained with linear regression is represented with the full red line. Linear regression is performed on the curve over the whole range of t$_e$ on the left and only over t$_e$ ranging from -45 to +45 on the right.

3 EXAMPLES OF APPLICATION – TH = 100 YEARS

The applicability of this method is tested on three examples for a time horizon of 100 years: i) the system studied in the previous chapter, and two examples from the literature: ii) energy production by solar panel and iii) emissions from a landfill.

3.1 FIRST EXAMPLE – SYSTEM FROM THE PREVIOUS CHAPTER

A first example is the case study used in chapter 3. The simplified temporal distributions representing the soil organic carbon changes during miscanthus growth are illustrated in Figure 24. The “linear” CO$_2$ emissions pattern over 15 years results in $[I_{dyn}/I_{stat}]_{CO2} = 1.08$ ($m(t_{min}) = 7500\ kg_{CO2}$). For the averaged inventory, emissions are symmetrical around t_0 and the $AGWP_{CO2}(100 - t_e)/AGWP_{CO2}(100)$ curve can be approximated by a linear function for $t_e \in [-45, 45]$, see the previous section. Thus, for both points of view, dynamic modelling does not change significantly the results compared to static modelling.

For wood growth, CO$_2$ capture is first simplified into three linear functions see Figure 25 ($t_1 = -180\ yr$, $m(t_1) = 0\ kg_{CO2}$, $t_2 = -75\ yr$, $m(t_2) = -0.005\ kg_{CO2}$, $t_3 = -25\ yr$, $m(t_3) = -0.011\ kg_{CO2}$, $t_4 = 0\ yr$, $m(t_4) = -0.006\ kg_{CO2}$) which results in $[I_{dyn}/I_{stat}]_{CO2} = 1.42$. CO$_2$ capture could be further simplified as a simple linear pattern ($t_{min} = -180\ yr$, $m(t_{min}) = 0\ kg_{CO2}$, $t_{max} = 0\ yr$ and $m(t_{max}) = -0.009\ kg_{CO2}$) which results in $[I_{dyn}/I_{stat}]_{CO2} = 1.43$, suggesting that further investigation with dynamic LCA is recommended.

To compare to the results of Chapter 3, the pulse emission due to the rest of the lifecycle occurring at t_0 is added: around 0.6 kgCO$_2$ for EP$_{zero}$ and 1.1 kgCO$_2$ for EP$_{conv}$ (“Energy production” parameter, see section 3.4 of chapter 3 for the definition). This results in $[I_{dyn}/I_{stat}]_{CO2} = 2.8$ for EP_{zero} and $[I_{dyn}/I_{stat}]_{CO2} = -0.4$ for EP_{conv}. This corresponds to what is observed in Figure 19, $[I_{dyn}/I_{stat}]_{CO2} = 2.80$ for EP_{zero} and $[I_{dyn}/I_{stat}]_{CO2} = -0.34$ for EP_{conv}.

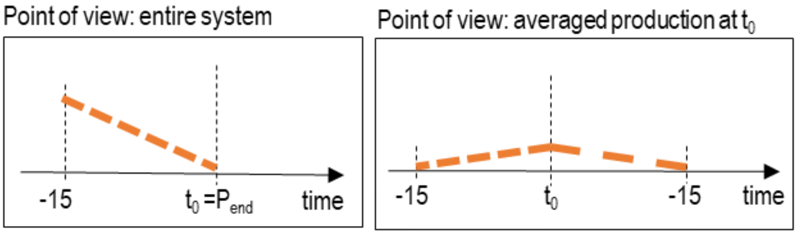

Figure 24: Simplified temporal distributions representing the soil organic carbon changes during miscanthus growth.

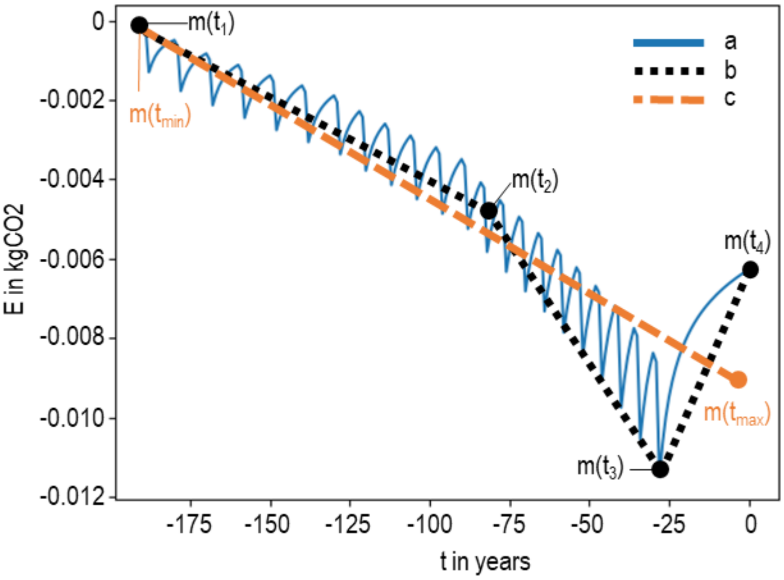

Figure 25: Temporal distributions representing CO_2 capture during the mean production of 1kg of wood residues at year 0 (plot with thinning). a: Temporal distribution calculated using Chapman-Richard equation and the algorithm for averaging a dynamic inventory (cf. Figure 15). b: First option to simplify the temporal distribution. c: Second option.

3.2 SECOND EXAMPLE – ENERGY PRODUCTION BY SOLAR PANEL

The following examples are taken from the literature. The Ecoinvent process “electricity production, photovoltaic, 570kWp open ground installation, multi-Si” (v3.10, geography FR), is interesting because the production of the infrastructure is the only contributor to the impact of this process on climate change. The impact is mainly due to fossil CO_2 emissions (GWP_{100} = 8.51x10^{-2} kgCO2eq, including 7.4x10^{-2} of fossil CO_2). The lifetime of the infrastructure is 30 years. To follow the recommendation of the previous chapter, P_{end} is set equal to t_0 resulting in the temporal distribution drawn in Figure 26. This results in $[I_{dyn}/I_{stat}]_{CO2} = 1.23$. In Figure 26 is also represented the averaged temporal distribution for electricity production at t_0, assuming constant production over 30 years. This leads to

$[I_{dyn}/I_{stat}]_{CO2} = 1.11$. If the uncertainty on static inventory data is lower than 20%, it is recommended to further investigate with dynamic LCA, particularly for a non-averaged point of view.

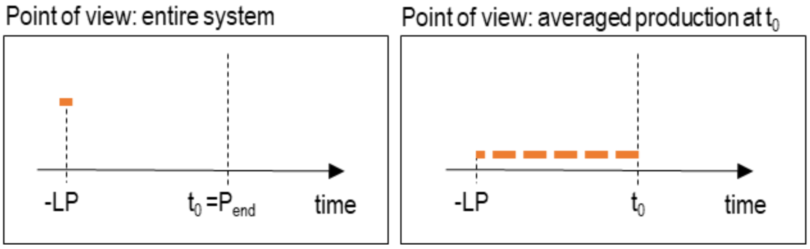

Figure 26: Temporal distributions representing the production of infrastructure for electricity generation with solar panels.

3.3 THIRD EXAMPLE – EMISSION FROM LANDFILL

Another interesting example is the case of landfills, where CH_4 emissions occur. The emissions pattern for CO_2 and CH_4 are inspired by the work of Wang (2020). The diversity of patterns is due to the different possible treatment of landfill gas: passive venting, flaring or energy recovery and the value of the decay rate. The types of simplified emissions patterns are summarised on Figure 27. The purple curve corresponds, for example, to a decay rate of 0.12 per year coupled with passive venting for CO_2 ($[I_{dyn}/I_{stat}]_{CO2} = 0.96$) and for CH_4 ($[I_{dyn}/I_{stat}]_{CH4} = 0.99$). The green curve corresponds, for example, to a decay rate of 0.04 per year coupled with energy recovery for CO_2 ($\left[\frac{I_{dyn}}{I_{stat}}\right]_{CO2} = 0.78$) and a decay rate of 0.04 per year coupled with passive venting for CH_4 ($[I_{dyn}/I_{stat}]_{CO2} = 0.97$). The blue curve corresponds, for example, to a decay rate of 0.02 per year coupled with flaring for CH4 ($[I_{dyn}/I_{stat}]_{CH4} = 0.84$). Not taking landfill dynamics into account could therefore lead to overestimating the impact on "climate change" at 100 years up to 20%.

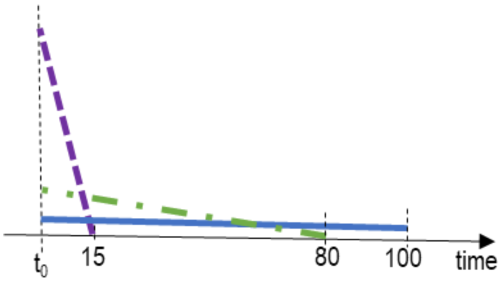

Figure 27: Types of simplified emissions patterns observed in Wang (2020) for the emissions of CO_2 and CH_4 in landfills

4 SETTING THRESHOLD

Four types of emissions patterns can be extracted from the examples and are listed in Figure 28. As demonstrated in section 1.1, if one of the extreme emission values of the "linear" emissions pattern is equal to zero, the mass ratio (and subsequently the $[I_{dyn}/I_{stat}]$ ratio) is independent of the value of the other extreme emission value. The mass ratio is also independent on the amplitude of the emission for "pulse" and "uniform" emissions pattern. Therefore, the $[I_{dyn}/I_{stat}]$ ratio can be plotted as a function of $t_{extreme}$ (i.e. t_{min} or t_{max} see Figure 28). In addition Figure 22 reveals that the all the $[I_{dyn}/I_{stat}]$ curves for $TH \in [100, 500]$ are contained between the $[I_{dyn}/I_{stat}]$ curves for $TH = 100\ and\ TH = 500$. To see the range of possibilities, the $[I_{dyn}/I_{stat}]$ is thus plotted for each of the four emissions patterns and for TH of 100 years and 500 years. The aim is to extract threshold values of pattern duration beyond which dynamic LCA is necessary to enable the LCA practitioner to identify if further investigation using dynamic LCA is necessary, using only two temporal information: the type of emissions pattern ("pulse", "linear with $m(t_{extreme}) = 0$", "linear with $m(t_0) = 0$", "uniform") and the pattern duration ($t_{extreme}$).

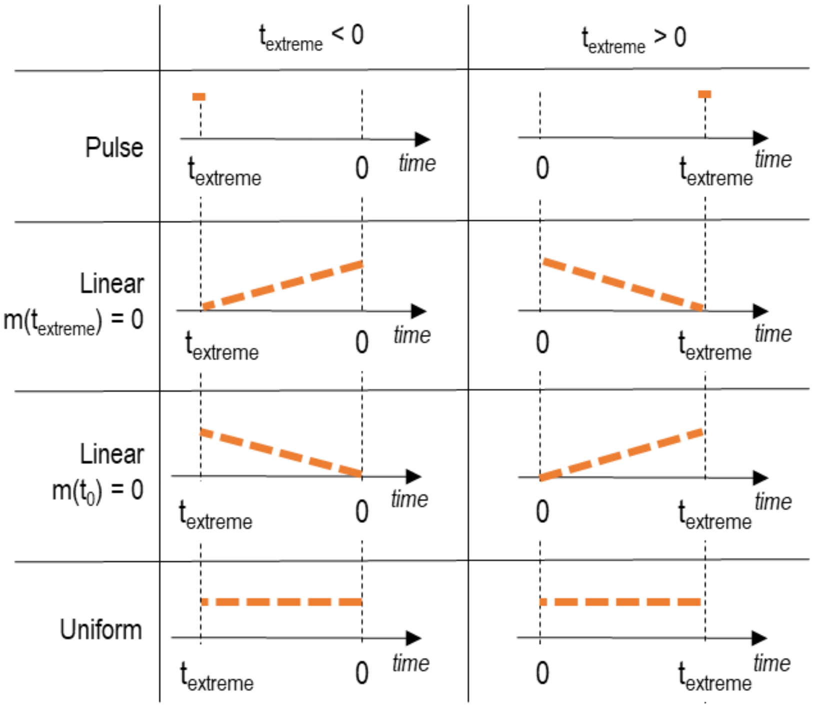

Figure 28: List of the emissions patterns extracted from the examples.

The commonly assumed uncertainty on climate change characterisation factor is 10% (Jolliet et al. 2015). Further investigation with dynamic LCA is deemed unnecessary if dynamic results differ less than 10% from static results. Figure 29 reveals that for CO_2, regardless of the chosen TH, further investigation with dynamic LCA is unnecessary if $t_{extreme}$ is included approximatively between:

- ± 13% of TH for a "pulse" emissions pattern. In the example on the solar panel, $t_{extreme}$ is equal to -30 years, dynamic LCA is necessary until a TH of 230 years (= -30/-13%).

- ± 40% of TH for a "linear with $m(t_{extreme}) = 0$" emissions pattern. In the example of landfills, $t_{extreme}$ is either 15 years, dynamic LCA is unnecessary, or $t_{extreme}$ is 80 years, dynamic LCA is necessary until a TH of 205 years. In the example of wood residues growth, $t_{extreme}$ is -180 years, dynamic LCA is necessary.
- ± 18% of TH for a "linear with $m(t_{extreme}) = 0$" emissions pattern.
- ± 26% of TH for a "uniform" emissions pattern. In the example of solar panel, $t_{extreme}$ is -30, dynamic LCA is necessary until a TH of 111 years.

Figure 30 reveals that for CH$_4$, regardless of the emissions pattern, if $TH \in [100, 500]$ and $t_{extreme} \in [-54\%\, TH, 47\%\, TH]$, further investigation with dynamic LCA is unnecessary. If $TH = 100$, further investigation with dynamic LCA is unnecessary for $t_{extreme}$ inferior to:

- 63% of TH for a "pulse" emissions pattern.
- 100% of TH for a "linear with $m(t_{extreme}) = 0$" emissions pattern. In the example of landfills, $t_{extreme}$ is either 15 or 80 years, dynamic LCA is unnecessary.
- 81% of TH for a "linear with $m(t_0) = 0$" emissions pattern.
- 91% of TH for a "uniform" emissions pattern. In the example of landfills, $t_{extreme}$ is 100 years, dynamic LCA is necessary.

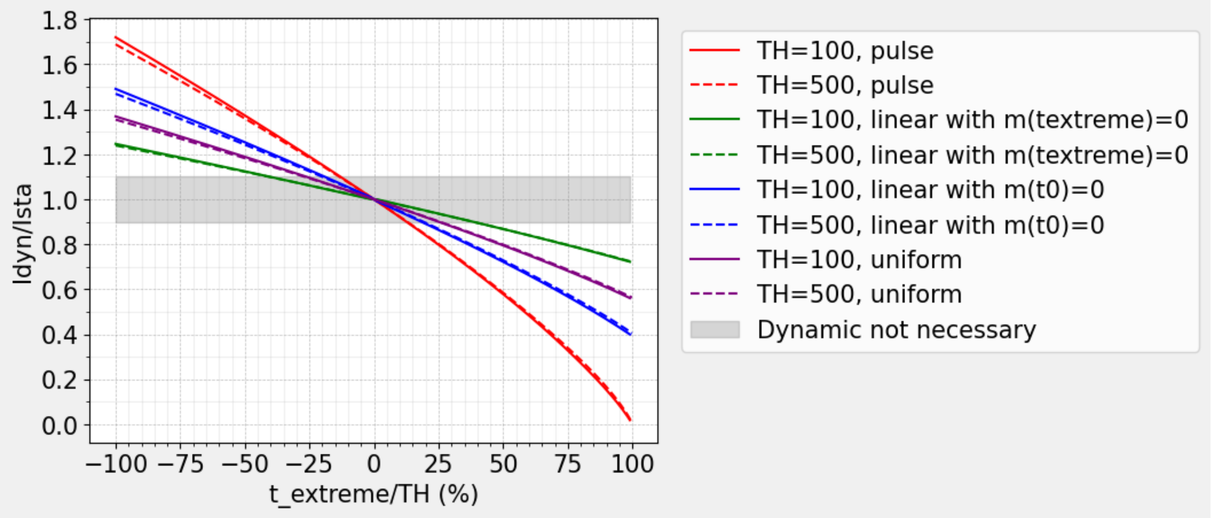

Figure 29: [I_{dyn}/I_{stat}] for CO$_2$ as a function of $t_{extreme}$ for the four emissions patterns defined in Figure 28 and two extreme value of TH: 100 and 500 years.

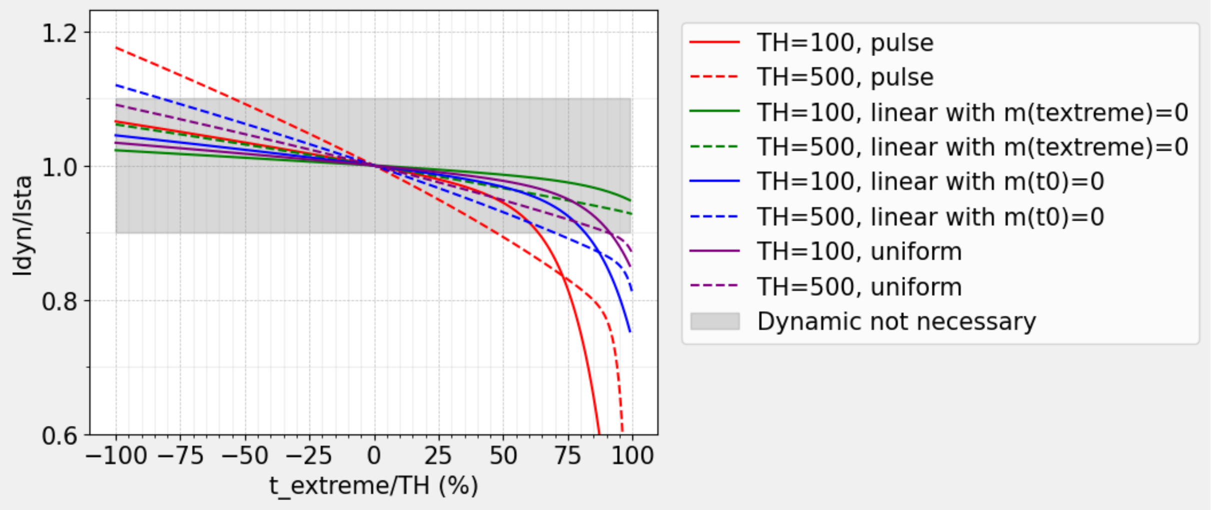

Figure 30: [I_{dyn}/I_{stat}] for CH_4 as a function of $t_{extreme}$ for the four emissions patterns defined in Figure 28 and two extreme value of TH: 100 and 500 years.

CHAPTER CONCLUSION

In this chapter, threshold values are provided, enabling the LCA practitioner to identify if further investigation using dynamic LCA is necessary, using only two temporal information: the type of emissions pattern ("pulse", "linear with $m(t_{extreme}) = 0$", "linear with $m(t_0) = 0$", "uniform") and the pattern duration ($t_{extreme}$). If the emissions pattern is more complex or if the LCA practitioner has further information on the uncertainty on static results and want to use a respectively lower or higher threshold to decide if dynamic LCA is necessary, the ratio $[I_{dyn}/I_{stat}]$ can be numerically calculated. These threshold values of pattern duration could be calculated for more GHG and for AGTP or other dynamic characterisation methods in future work.

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General conclusion et perspectives

This thesis seeks to answer the research question: How to address the methodological challenges associated with the LCA of CCUNET systems? Over the course of the four chapters, methodological and practical contributions have been made to facilitate and improve the assessment of negative emissions of CCU systems, as detailed in section 1. Shortcomings in the scientific literature have been identified throughout this work, as described in section 2, related to inventory data and impact characterisation. In the last subsection (2.3), a reflection on LCA as a decision-making tool in the context of CCU or NET systems is presented.

1 RESULTS SUMMARY

This thesis seeks to answer the research question: How to address the methodological challenges associated with the LCA of CCUNET systems? Over the course of the four chapters, methodological and practical contributions have been made to facilitate and improve the assessment of negative emissions of CCU systems, as detailed in the first and second subsection respectively.

1.1 METHODOLOGICAL CONTRIBUTIONS

This thesis formulates recommendations for harmonising practices in LCA of CCUNET systems, in addition to the existing recommendations for CCU systems. The proposed recommendations in chapter 2 for evaluating the negative emissions potential of a CCUNET system are to carry out an attributional LCA, use system extension to solve multifunctionality, and count atmospheric CO_2 using the "+1/-1" approach. Evaluating negative emissions potential falls within the definition of an attributional goal (cf. chapter 2, section 4.4.2). System extension allows for flows of atmospheric carbon to be monitored from CO_2 capture to release without distortion (cf. chapter 2, section 4.2). The "+1/-1" approach is a requirement to perform dynamic LCA (cf. chapter 2, section 4.3). Using the method described in chapter 4, dynamic LCA should be used if the $[I_{dyn}/I_{stat}]$ ratio of one of the GHGs contributing most to the impact is greater than 1.1 or less than 0.9 (default difference of 10%, to be adapted according to uncertainty of the static inventory data). The concept of negative emissions has emerged in response to the climate objective of carbon neutrality by 2050. If a dynamic LCA is indeed required to provide a more precise answer to the question "does the system generate negative emissions?", we therefore recommend using the functional unit "total amount of units produced over the entire lifespan of the system" and calculating results for calendar-based time horizons (cf. chapter 3, section 5.2). Moreover, to compare CCUNET to other NET systems, it is suggested to use the "treatment of atmospheric CO_2" functional unit as the common function between systems (cf. chapter 2, section 4.1). In that case, if a dynamic LCA is required, it is then preferable to use the functional unit "production of several units of the product or service at t_0" to smooth out the differences in the temporal distribution of production between the compared systems (cf. chapter 3, section 5.2).

Chapter 3 and chapter 4 contribute to the methodological development of dynamic LCA for climate change. In Chapter 3, an algorithm is proposed for harmonising the transformation of an inventory modelling the system over its entire lifetime into an average product-oriented inventory (production of one unit at a given time)(cf. Figure 15). The relationship between the inventory timeline and the impact characterisation timeline is also clarified to enable a consistent use of time horizons across LCA studies (cf. chapter 3, section 3.3). This results in recommendations on the definition of the functional unit (cf. chapter 3, section 6):

- the “total amount of units produced over the entire lifespan of the plant” functional unit should be used for evaluating the potential impact on climate change of the entire system relative to climate goals on a calendar-based timeline. To compare the obtained results to static results, the time horizon should be defined starting from the last year of production.
- the “1 unit produced at t_0” functional unit should be used for comparing systems that do not share the same temporal distribution of production and to build inventory data reusable as background inventory data in another life cycle.

In chapter 4, a methodology is presented to enable the LCA practitioner to assess, using only simplified temporal information, whether dynamic or static LCA should be performed. This makes it possible to focus efforts more effectively and potentially save time for improving static inventory data.

1.2 PRACTICAL CONTRIBUTIONS

Firstly, generic inventory data are proposed in chapter 1 and chapter 2 to facilitate the use of the "from-cradle-to-grave" system boundaries during the evaluation of CCUNET systems. The CO_2 source and the end-of-life of the CO_2-based products are often not modelled in LCA of CCU systems. BECCS systems are both promising CO_2 sources and end-of-life solutions, in order to generate negative emissions. Therefore, in chapter 1 (section 3.3), generic LCI are provided to easily model steps of BECCS systems: biomass harvesting, pretreatment, combustion, gasification, CO_2 capture with a monoethanolamine based solvent, CO_2 transport by pipelines and storage in a geological reservoir. Growing biomass is an efficient way of capturing CO_2 from the atmosphere. The by-products of biomass transformation, such as the distiller's dried drains with soluble in the case of maize fermentation, can be valorised as animal feed. In this case, to be able to use the recommended “+1/-1” approach to account for atmospheric CO_2 and "from-cradle-to-grave" system boundaries, a conversion factor is proposed in chapter 2 (section 2.3) to convert the mass of carbon ingested by ruminants into kilograms of CO_2 and CH_4 released back into the atmosphere. Hence, for 1 kg of carbon ingested, around 0.7 kg is emitted as CO_2 and 0.3 kg as CH_4. Chapter 1 is also a reminder that the adequacy of the quality of the inventory data in relation to the LCA objective must be studied. The generic inventory data proposed in this thesis are adapted to carry out the first iteration of a LCA. If a first iteration reveals a strong contribution of these inventories to the results, they ought to be further studied and potentially improved by process modelling or onsite measurements.

Secondly, LCA were carried out using emerging tools in chapter 2 and chapter 3, i.e. Brightway2 and its Temporalis package. All the documents created to generate the results (excels, jupyter notebook), as well as the exact version of the various python packages and databases used, are provided as supplementary information to the articles allowing anyone to reproduce the results. Brightway2 allows the creation of the inventory by importing LCI data stored in an Excel sheet. Sharing an excel file with dedicated sheets for the import in Brightway2 framework) is used to carry out dynamic LCA, increasing the understanding of this tool. An improved version will be provided as supplementary information to the article presented in chapter 3. The practicality of Temporalis for performing a dynamic LCA is demonstrated and examples are provided to facilitate future dynamic LCAs with this tool. These examples notably include:

- a proposal to include temporal information into the LCI excel sheet,
- the construction of temporal distribution for CO_2 capture during the growth of wood residues on a plot where thinning is carried out using the Chapman Richard formula,
- examples of results visualisation (impact and inventory).

2 PERSPECTIVES

Shortcomings in the scientific literature have been identified throughout this work. They can be grouped into two main categories: shortcomings related to inventory data (6.2.1), and shortcomings related to impact characterisation (6.2.2). In a third subsection (6.2.3), a reflection on LCA as a decision-making tool in the context of CCU or NET systems is presented.

2.1 RESEARCH GAPS AND PERSPECTIVES: INVENTORY DATA

LCA is a data-driven methodology. Chapter 1 highlights that further work is needed to generate robust, complete and up-to-date data for modelling every BECCS systems. For instance, the energy requirements for biomass harvesting (before 2016), the bed material consumption for gasification (before 2011), or the consumption of activated carbon and NaOH in MEA-based capture (before 2006) need to be updated (cf. chapter 1 section 4.1.1). Chapter 1 (section 4.2.2) also revealed that too few studies have been carried out on stand-alone anaerobic fermentations, second-generation CO_2 capture processes or other types of CO_2 transport (i.e. rail) and storage options. In addition, the recommendation in chapter 2 to use the “+1/-1” approach to account for atmospheric CO_2 means that the knowledge on the fate of carbon during food and feed steps needs to be improved. And lastly, in chapter 3, the lack of a dynamic background LCI database was recalled.

Collecting and sharing LCI data is an area of research in LCA, as illustrated by the review of Saavedra-Rubio (2022). They are in line with the findings of chapter 1: there is a lack of transparency regarding LCI data and their collection methods. They proposed a template for harmonising the collection of inventory data and encouraging its sharing. This type of approach could be taken a step further by adding to their excel template a sheet that can be directly imported into Brightway2 (including essential information such as the version of Brightway2 used or the version of the databases for instance), with dedicated fields to indicate temporal data for dynamic LCA. A widespread use of such template could be the foundation for the creation of a LCI database by and for Brightway2 users along with the workflow proposed by Ghose (2024) for FAIR inventory data (Findable, Accessible, Interoperable, Reusable). The spreadsheet format is suitable for sharing FAIR data as it is easy to open and can even be reused without Brightway2 by manually transforming the data (for example by entering it into Simapro). The assessment of data quality should be the responsibility of the data user, who must assess it in relation to his own LCA objectives, which may differ from the ones of the data provider.

The purpose of compiling an LCI to model a system is to evaluate the potential impact of the system on the environment and decide whether it is worth to deploy it. This decision must be based on results consistent with the territory in which the system is to be deployed. Actually, the French low carbon national strategy (MTES 2020) indicates that regions and intercommunal bodies are key players for the practical implementation of climate policies. Many of the contributors to the impact of a CCUNET system are specific to the territory in which the system is to be deployed: biomass, energy, transportation distances, or even the products end-of-life which will depend on the infrastructure available in the region. The contextualisation of inventory data within a territory is still a field of research in LCA, as illustrated by the work of Loiseau (2021) on territorial LCA. For instance, there are still questions about how to take account of consequential effects within territorial LCA. For CCU system, consequential effects include indirect land use change and rebound effects due to a potential increase of economic activity (Haut conseil pour le climat 2023). Assessing the system within a given territory is also an opportunity to question its usefulness relative to the real needs of the territory.

The research perspectives regarding inventory data are thus:

- to apply the workflow proposed by Ghose (2024) to the inventories created by Brightway2 users in order to build an open-source database for a more efficient data sharing
- improve the assessment of potential negative emissions from CCUNET systems by using territorial LCA, including consequential effects and coupling it with dynamic LCIA if relevant.

2.2 RESEARCH GAP AND PERSPECTIVES: IMPACT CHARACTERISATION ON CLIMATE CHANGE

In this work two assumptions are made in the AGWP formulas (and corresponding parameters) proposed in the fifth assessment report of the IPCC (IPCC 2013): a symmetry of response of the climate system to the uptake and release of CO_2 and a constant CO_2 background concentration. Therefore, several avenues of research exist to improve the characterisation of the impact on climate change. The symmetry assumption could be checked, and if not correct, a specific formula should be proposed to characterise the impact of an uptake of CO_2. Concerning the CO_2 background concentration, impulse response function of CO_2 (IRF) could be calculated for each representative concentration pathways (RCP) proposed by the IPCC. To be thorough, the IRF also depends on the year of emission and its associated CO_2 background concentration. IRF could thus be calculated for each RCP and for a variety of years of emissions. The step between the year of emissions would need to be defined to find the right balance between precision and complexity of the characterisation step. However, it would increase the dependence of the calculated results on subjective choices (Levasseur et al. 2016). As indicated in chapter 3, a more general examination of how to account for the uncertainty of the characterisation factors in LCA seems more useful to address this issue.

Improving the characterisation step in Temporalis in Chapter 4 provided an opportunity to examine the parameters given by the IPCC in greater detail and to become aware of the sources of variability in the AGWP formulas. The IPCC provides uncertainty for each parameter used in the AGWP formula: radiative efficiency, perturbation lifetime, indirect effects and so on. Another source of uncertainty in the calculation of the AGWP is the effect of carbon feedback on climate change. Emissions of GHG into the atmosphere induce global warming, which in turn reduce the efficiency of the natural carbon sinks. This results in an increase of the CO_2 concentration in the atmosphere, i.e. climate carbon feedback. For now, the values of GWP for 20, 100 and 500 years are given with and without climate carbon feedback but not the parameters of the formulas. This means that climate carbon feedback cannot be included in dynamic LCIA. Another example of a source of variability is the modelling of the oxidation of methane into CO_2. It can vary between 50% of the carbon oxidised as CO_2 and 100%.

Bamber et al. (2020) indicate that less than 20% of LCA studies published since 2014 contain a qualitative or quantitative analysis of the results uncertainty. The study of uncertainties, or at least its communication, within an LCA article is therefore not a common practice. Lo Piano and Benini (2022) reviewed approaches for uncertainty appraisal in LCA. Only 20% of the articles they reviewed carried out an uncertainty study on characterisation factors. They observed that uncertainty studies on characterisation factors for climate change focus on the chosen time horizon, static vs. dynamic, and land use change. Both Bamber et al. (2020) and Lo Piano and Benini (2022) encourage the evaluation and communication of uncertainties so that LCA remains a credible decision-making tool. Bamber et al. (2020) observed that Monte Carlo analysis is widely used to propagate uncertainties because it is available in common LCA software, even though it is not necessarily the most appropriate method. This shows the importance of promoting good practice and integrating several methods for managing uncertainty into LCA software. Including uncertainties due to the characterisation step in LCA studies is still a challenge on two aspects: ensuring that it is correctly performed by the LCA practitioner and clearly communicating the results.

The research perspectives regarding climate change impact characterisation are thus:

- to check the assumption of a symmetry of the climate system to uptake and release of CO_2,
- to include climate carbon feedback in dynamic LCA,
- to find a way to include uncertainty on the characterisation method in Temporalis while keeping results clarity.

2.3 RESEARCH GAP AND PERSPECTIVES: LCA IN DECISION-MAKING

The majority of CCU and NET technologies have low technological maturity. CCU technologies are developed with the aim of reducing CO_2 emissions and fossil-resource consumption (Ramirez et al. 2020). NET technologies are developed for reducing the atmospheric concentration of CO_2. Therefore, verifying the environmental relevance of a CCU or NET technology should be at the centre of the technology design, even if this poses challenges of data availability and scaling, from lab scale to industrial scale. Using "from-cradle-to-grave" boundaries to evaluate the environmental performance of a CCU or NET technology at the concept stage of its design allows questioning the final utility of its products. For instance, the case study chosen in this work (reusable bag) is not really compatible with an idea of sufficiency. However, some uses of plastics will remain essential, such as medical use (e.g., gloves). Using "from-cradle-to-grave" boundaries, and system expansion to deal with multifunctionality, also allow to identify if the technology will actually generate negative emissions or if reaching negative emissions will be dependent on external factors not controllable by the technology developer such as its usage or the end-of-life of its products (ex: CO_2-based methanol can be used as a fuel (no negative emissions) or a raw material for plastic production (potential negative emissions)).

Accounting for the impact on climate change using LCA may also be a legal necessity (e.g., Renewable Energy Directive (European Parliament and Council 2018)) or a tool to obtain subsidies (e.g., low-carbon label France (MTECT 2024a)), or for communication (e.g., PEF (European Commission 2021)). Two regulatory obligations are gaining momentum in Europe: environmental labelling (ADEME 2024) and EU carbon market (cap and trade principle) (MTECT 2024b). In both cases, the concern raised by Tanzer and Ramirez (2019), about promoting solutions under the guise of negative emissions when they are not, applies. For environmental labelling, system expansion cannot be used to deal with multifunctionality. How to distribute impacts so that if a system does not generate negative emissions, none of its products will score negatively? For the EU carbon market, Europe has recently proposed a provisional agreement to certify negative emissions (European Parliament and Council 2024). Negative emissions should be calculated with “from-cradle-to-grave” system boundaries: how to distribute the certified negative emissions between the different actors of the life cycle (notably between the CO_2 source, the CO_2 capture, the CO_2 permanent storage)?

The desire to produce negative emissions is intrinsically linked to the objective of carbon neutrality by 2050. LCA can then be a tool to help public decision-makers build relevant net-zero strategies for their territory. Guérin-Schneider et al. (2018) have demonstrated that the creation of a tool, enabling non-specialists to carry out an LCA of a water treatment system, has enabled the appropriation of LCA by local authorities. Following their feedback and recommendations, a similar tool could be created for NET and CCU solutions with an additional dynamic dimension to relate the LCA results on climate change to the precise timeline of climate goals. Such a tool should contain general inventory data to describe existing processes for biomass growth, biomass transformation, CO_2 capture, transport, storage, transformation and so on, and include territory-specific parameters such as the transportation distance, the initial stock of carbon in the soil or the energy production technologies.

The research perspectives regarding LCA in decision-making, to help decide on the best technical options of CCUNET systems to develop and/or finance, are thus:

- to translate the recommendations to the specific framework of carbon accounting (e.g. environmental labelling, EU carbon market)
- to build a dynamic territorial LCA tool to facilitate the appropriation of CCU/NET technologies by public decision-makers.

If we do not change our current rate of emissions, the planetary boundary for climate change will be exceeded around 2035: it is urgent to take action. CCUNET technologies have the potential to accelerate the transition towards carbon neutrality, but the primary lever for action is to change our way of life in order to build a more sufficient society. LCA is an essential tool that should be put in the hands of stakeholders to plan a transition that limits impacts transfers both between categories (acidification, eutrophication, etc.) and over time (current vs. future generations).

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Annexes

1 CIRCULAR FOOTPRINT FORMULA

In the guidelines proposed by the JRC (Nessi et al. 2021), the CO_2 is a waste to be treated. The JRC recommends the use of the CFF. The CFF was developed to allocate the burdens and credits of recycling between supplier of waste and user of recycled raw materials. It is divided in three parts, material recovery, energy recovery and disposal. The formula for material recovery is shown on Figure 31. The CFF is built in comparison to a baseline scenario where no recycled material is used. This way the sum of the impact of the supplier life cycle and the impact of the user life cycle is equal to the impact obtained with system expansion. The formula includes the impact due to the production of primary material in the baseline scenario:

$$\left((1-R_1)+R_1\frac{Q_{sin}}{Q_p}\right)E_v \tag{1}$$

The impact and credit due to the amount of primary material production avoided by using recycled material is then allocated to the user with the allocation factor A:

$$A\left(R_1E_{recycled}-R_1E_v\frac{Q_{sin}}{Q_p}\right) \tag{2}$$

Adding and factoring these two terms (equations 1 and 2) gives the first part of the CFF for material recovery:

$$(1-\mathrm{R}_1)\mathrm{E}_\mathrm{v}+\mathrm{R}_1\left(\mathrm{AE}_\mathrm{recycled}+(1-\mathrm{A})\frac{\mathrm{Q}_\mathrm{sin}}{\mathrm{Q}_\mathrm{p}}\mathrm{E}_\mathrm{v}\right) \tag{3}$$

Following the same reasoning, the impact and credit due to the amount of primary material production avoided by recycling material in the EoL is allocated to the supplier with the allocation factor $(1-A)$:

$$(1-A)R_2*\left(E_{recyclingEoL}-\frac{Q_{sout}}{Q_p}E_v^*\right) \tag{4}$$

It corresponds to the second part of the CFF for material recovery. $\frac{Q_{sin}}{Q_p}$ and $\frac{Q_{sout}}{Q_p}$ enables to include the difference in quality between recycled material and primary material.

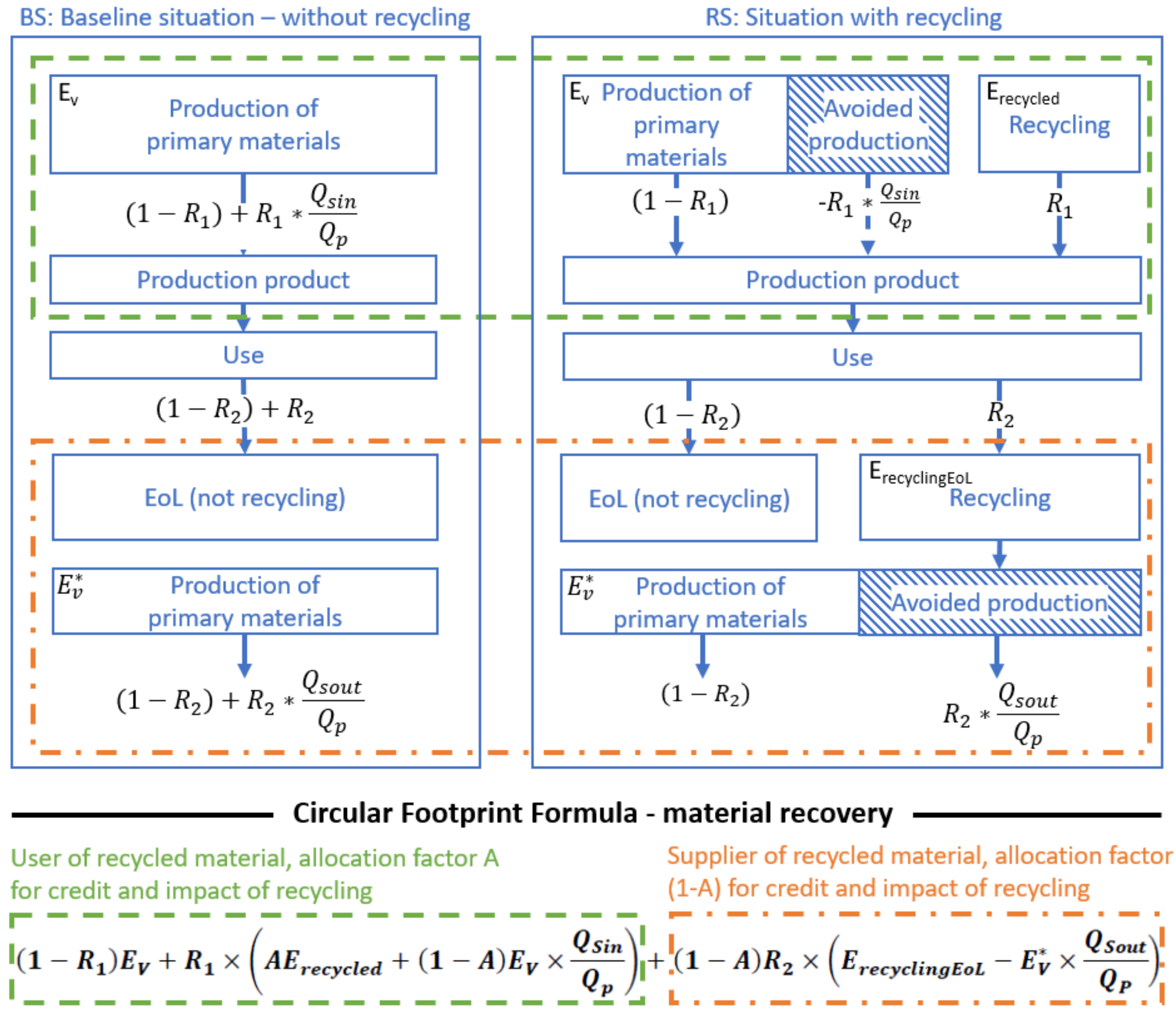

Figure 31: Circular Footprint Formula for material recovery copied from the section 4.4.10.2 of the JRC guidelines for LCA of plastics from alternative feedstocks (Nessi et al. 2021) and explanation of the terms

In the CCUNET case study, only the first part of the CFF for material recovery is needed. The point of substitution is defined as the moment when the process becomes identical between the CO_2-based product and the fossil-based product (Nessi et al. 2021). In the CCUNET case study, the point of substitution is reached when propylene is produced by MTO. Therefore, the production of primary material corresponds to the production of fossil-based propylene. The quality ratio is assumed to be 1, i.e. the CO_2-based propylene has the same quality as fossil-based propylene. The allocation factor A between supplier and user is assumed to be 0.5 for two reasons: i) there is currently no established market for CO_2-based propylene, ii) the default value for PP proposed in appendix C of the Plastic LCA method is 0.5 (Nessi et al. 2021; Nessi et al. 2022). However, as pointed out by the JRC, “any future study focusing on real products shall take into account the specific market situation at the time of the study itself.” The CO_2 is considered as a waste. It means that none of the impacts arising from the activities before CO_2 capture are attributed to the CO_2-based product. It raises an issue: the mass balance between the amount of CO_2 sequestered by photosynthesis and the amount of CO_2 captured for use is not kept. It raises a challenge on the interpretation of the results.

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2 MATHEMATICAL DEMONSTRATIONS FOR CHAPTER 3

The radiative efficiency of $\mathrm{CO_2}$ is noted a_{CO2}. The decay function of $\mathrm{CO_2}$ into the atmosphere is noted C_{CO2}, with τ_i and a_i being the parameters defined in Joos et al. (Joos et al. 2013). A simplified system emitting only $\mathrm{CO_2}$ is studied. m_e is the mass emitted at time te.

$$\boldsymbol{\lim_{TH\to+\infty}\frac{I_{FU1(P_0,TH)}+I_{FU1(P_{end},TH)}}{2}=\lim_{TH\to+\infty}I_{FU2(TH)}}$$

- TH: time horizon
- $I_{FU1(P_0,TH)}$: Impact calculated using the “20000 bags produced over the lifespan of the plant” functional unit with the dynamic inventory positioned so that $P_0=t_0$
- $I_{FU1(P_{end},TH)}$: Impact calculated using the “20000 bags produced over the lifespan of the plant” functional unit with the dynamic inventory positioned so that $P_{end}=t_0$
- $P_{end}-P_0=LP$:lifespan of the plant
- $I_{FU2(TH)}$: Impact calculated using the “20000 bags produced at t0” functional unit

Demonstration:

Let’s look at the first part of the equation:

$$\frac{I_{FU1(P_0,TH)}+I_{FU1(P_{end},TH)}}{2}=\frac{a_{CO2}}{2}\left[\sum_{te=0=P_0}^{LP}m_e\int_0^{TH-te}C_{CO2}(t)dt+\sum_{te=-LP}^{0=P_{end}}m_e\int_0^{TH-te}C_{CO2}(t)dt\right]\tag{1.1}$$

$$\frac{I_{FU1(P_0,TH)}+I_{FU1(P_{end},TH)}}{2}=\frac{a_{CO2}}{2}\left[\sum_{te=0=P_0}^{LP}m_e\int_0^{TH-te}C_{CO2}(t)dt+\sum_{te=0}^{LP}m_e\int_0^{TH-(te-LP)}C_{CO2}(t)dt\right]\tag{1.2}$$

The system studied is identical between $FU1(P_0,TH)$ and $FU1(P_{end},TH)$: the masses m_e are identically distributed over time.

$$\frac{I_{FU1(P_0,TH)}+I_{FU1(P_{end},TH)}}{2}=\frac{a_{CO2}}{2}\sum_{te}m_e\left[\int_0^{TH-te}C_{CO2}(t)dt+\int_0^{TH+LP-te}C_{CO2}(t)dt\right]\tag{1.3}$$

$$\frac{I_{FU1(P_0,TH)}+I_{FU1(P_{end},TH)}}{2}=\frac{a_{CO2}}{2}\sum_{te}m_e\left[2\int_0^{TH-te}C_{CO2}(t)dt+\int_{TH-te}^{TH+LP-te}C_{CO2}(t)dt\right]\tag{1.4}$$

$$\lim_{TH\to+\infty}\left(\frac{I_{FU1(P_0,TH)}+I_{FU1(P_{end},TH)}}{2}\right)=Ma_{CO2}\left[a_0\left[TH+\frac{LP}{2}\right]+\sum_{i=1}^{3}a_i\tau_i\right]-a_{CO2}a_0\sum_{te}m_e te\tag{1.5}$$

$$\lim_{TH\to+\infty}\left(\frac{I_{FU1(P_0,TH)}+I_{FU1(P_{end},TH)}}{2}\right)=Ma_{CO2}\left[a_0\left[TH+\frac{LP}{2}\right]+\sum_{i=1}^{3}a_i\tau_i\right]-a_{CO2}a_0\frac{LP}{2}M\tag{1.6}$$

$$\lim_{TH\to+\infty}\left(\frac{I_{FU1(P_0,TH)}+I_{FU1(P_{end},TH)}}{2}\right)=Ma_{CO2}\left[a_0TH+\sum_{i=1}^{3}a_i\tau_i\right]=\lim_{TH\to+\infty}\left(I_{FU2(TH)}\right)\qquad(1.7)$$

Let's look at the second part of the equation:

$$I_{FU2(TH)}=M\int_0^{TH}a_{CO2}C_{CO2}(t)dt\qquad(2.1)$$

$$\lim_{TH\to+\infty}\left(I_{FU2(TH)}\right)=Ma_{CO2}\left[a_0TH+\sum_{i=1}^{3}a_i\tau_i\right]\qquad(2.2)$$

The equality is verified.

$$I_{FU1(P_0,TH_1)}=I_{FU1(P_{end},TH_3)}=I_{FU2(TH_2)}\xRightarrow[TH\to+\infty]{}TH_2=TH_1-\frac{LP}{2}=TH_3+\frac{LP}{2}$$

- TH_1, TH_2, TH_3: three different time horizons
- $I_{FU1(P_0,TH)}$: Impact calculated using the "20000 bags produced over the lifespan of the plant" functional unit with the dynamic inventory positioned so that $P_0=t_0$
- $I_{FU1(P_{end},TH)}$: Impact calculated using the "20000 bags produced over the lifespan of the plant" functional unit with the dynamic inventory positioned so that $P_{end}=t_0$
- $P_{end}-P_0=LP$: lifespan of the plant
- $I_{FU2(TH)}$: Impact calculated using the "20000 bags produced at t0" functional unit

Demonstration:

$$\lim_{TH1\to+\infty}\left(I_{FU1(P_0,TH_1)}\right)=a_{CO2}a_0\left[M(TH_1)-\sum_{te=0=P_0}^{LP}\frac{M}{LP+1}*t_e\right]+\sum_{i=1}^{3}a_i\tau_i\qquad(3.1)$$

$$\lim_{TH1\to+\infty}\left(I_{FU1(P_0,TH_1)}\right)=a_{CO2}a_0\left[M(TH_1)-M\frac{LP}{2}\right]+\sum_{i=1}^{3}a_i\tau_i\qquad(3.2)$$

$$\lim_{TH3\to+\infty}\left(I_{FU1(P_{end},TH_3)}\right)=a_{CO2}a_0\left[M(TH_3)-\sum_{te=-LP}^{0=P_{end}}\frac{M}{LP+1}*t_e\right]+\sum_{i=1}^{3}a_i\tau_i\qquad(4.1)$$

$$\lim_{TH3\to+\infty}\left(I_{FU1(P_{end},TH_3)}\right)=a_{CO2}a_0\left[M(TH_3)+M\frac{LP}{2}\right]+\sum_{i=1}^{3}a_i\tau_i\qquad(4.2)$$

$$\lim_{TH2\to +\infty}\left(I_{FU2_{(TH2)}}\right) = a_{CO2}a_0MTH_2 + \sum_{i=1}^{3} a_i\tau_i \tag{5.1}$$

Using Equation (3.2), (4.1) and (5.1):

$$I_{FU1(P_0,TH_1)} = I_{FU1(P_{end},TH_3)} = I_{FU2(TH_2)} \xRightarrow[TH\to +\infty]{} TH_2 = TH_1 - \frac{LP}{2} = TH_3 + \frac{LP}{2}$$

$$\lim_{TH\to +\infty}\left(I_{static} - I_{dynamic}\right) = a_{CO2}a_0\sum_{te} m_e t_e$$

- I_{static} : impact calculated with the static approach (all emissions occur at t_0)
- $I_{dynamic}$: impact calculated with the dynamic approach

Demonstration:

$$\lim_{TH\to +\infty}\left(I_{static} - I_{dynamic}\right) = a_{CO2}a_0\sum_{te} m_e t_e$$

$$I_{static} - I_{dynamic} = \sum_{te} m_e a_{CO2}\left(\int_0^{TH} C_{CO2}(t)dt - \int_0^{TH-te} C_{CO2}(t)dt\right)$$

$$I_{static} - I_{dynamic} = \sum_{te} m_e a_{CO2}\left(\int_{TH-te}^{TH} C_{CO2}(t)dt\right)$$

$$I_{static} - I_{dynamic} = \sum_{te} m_e a_{CO2_0}[a_0 t_e + \sum_{i=1}^{3} a_i\tau_i[exp\left(-\frac{TH-te}{\tau_i}\right) - exp\left(-\frac{TH}{\tau_i}\right)]]$$

$$\lim_{TH\to +\infty}\left(I_{static} - I_{dynamic}\right) = a_{CO2}a_0\sum_{te} m_e t_e$$

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3 MISCANTHUS: MODELLING BIOGENIC CO_2 CAPTURE AND EMISSIONS DURING BIOMASS PRODUCTION (PLANT AND SOIL)

The most basic approach to evaluate the quantity of CO_2 captured ($A_{CO2,captured}$, kg_{CO2}) during biomass growth is to use the carbon content (x_c, $kg_C/kg_{product}$) and the mass (m, $kg_{product}$) of the biomass-based product. Knowing the molar mass of CO_2 (M_{CO2}, kg_{CO2}/mol) and the molar mass of carbon (M_C kg_C/mol), the mass of the biomass-based product is converted to a mass of CO_2 captured following the equation:

$$A_{CO2,captured} = m x_C \frac{M_{CO2}}{M_C} \tag{1}$$

However, the biomass-based products are only one carbon pool of the biomass production system. Carbon is also stored in the roots and in the soil. In fact 5 to 15% of global fossil fuel emissions could be offset by SOC sequestration (Goglio et al. 2015). This potential of SOC sequestration has also been recognised during the COP21 with the launch of the “4 per 1000” research programme (Ministère de l'agriculture et de la souveraineté alimentaire 2015). This programme aims at increasing the SOC stock by 4 per 1000 per year. Hence the importance of taking into account SOC in the environmental assessment of the biomass production system. Goglio et al. (Goglio et al. 2015) conducted a comprehensive study of available models to include SOC variations in LCI. They did not identify a consensus on the method to be used. This lack of consensus is still currently valid as observed in the literature review performed on LCI of BECCS systems.

Goglio et al. (Goglio et al. 2015) identified four types of methods to calculate SOC changes (listed in increasing order of preference, as given in their recommendation):

- “Emission factor estimation methods” (e.g. IPCC Tier I factors (IPCC)) correspond to the use of default emissions factors that depend on few parameters (e.g. the climate or crop management). These emission factors are easy to use but do not allow to consider temporal dynamics of SOC variations.
- “Simple carbon models” are based on few simple equations and do not include modelling of biomass growth. The time step is in general a year. These models are easy to use thank to their low data requirement.
- “Dynamic crop-climate-soil models” are more complex mathematical models as they aim to describe the whole system, i.e. the interactions between biomass growth, environmental processes and SOC stock variations. The time step is in general a day. These models are data

intensive and require expertise to operate. These models are thus difficult to use to perform an LCA.

- “Measurement” is costly and time consuming to perform.

Due to data availability and degree of expertise on crop-climate-soil models, “Simple carbon models” will be used to model the SOC stock variations in this thesis. In her PhD thesis, Ariane Albers (Ariane Albers 2019) identified the ICBM (Introductory Carbon Balance Model) (Andrén and Kätterer 1997) and AMG models (Clivot et al. 2019) as promising to include temporal dynamics of SOC stock changes while keeping a low data requirement. Both the ICBM model (Andrén and Kätterer 1997) and the AMG model (Clivot et al. 2019) follow the approach proposed by Hénin and Dupuis (Hénin and Dupuis 1945). The difference between the two models is mainly due to the geographical origin of the data used for the model parameterisation: Sweden for the ICBM model (Andrén and Kätterer 1997), France for AMG (Clivot et al. 2019). Moreover, since 2012, AMG is used in a simulation tool for farmers and others parties concerned with carbon storage in agricultural soils (AgroTransfert 2019). AMG is thus undergoing continuous improvement. As AMG is an up-to-date and applicable model for the geography of our case study, AMG is selected to model the SOC stock variation due to maize and miscanthus productions.

AMG is based on the two following equations:

$$QC = QC_S + QC_A \tag{2}$$

$$\frac{dQC_A}{dt} = \sum_i m_i h_i - kQC_A \tag{3}$$

with:

- QC: the total stock of SOC (t.ha^{-1}),
- QC_S: the stable fraction of the SOC (t.ha^{-1}),
- QC_A: the active fraction of the SOC (t.ha^{-1}),
- i: a type of organic carbon source (agricultural residues, soil amendments),
- m_i: annual input of organic carbon (t.ha^{-1}.yr^{-1}),
- h_i: isohumic coefficient (unitless),
- k: mineralisation constant (yr^{-1}).

The parameters of interest to model the case study are summarised in Table 14 and are mainly taken from the parameters collected by Ariane Albers (Ariane Albers 2019) during her PhD thesis. Albers assumed no LUC. With this assumption, the stable fraction of the SOC can be assumed constant and equal to 48.04 tC.ha^{-1}. The initial stock of SOC is assumed to be 50 tC.ha^{-1}. It corresponds to a ratio of stable carbon to total carbon equal to 96%. Clivot *et al.* rather assumed a ratio of 65%. The climate is a "degraded oceanic climate of Central and Northern Plains", i.e. the climate of a large area of cereals and oil crops in France according to Albers. However, the climate in France seems to be "warm temperate moist" in Figure 3A.5.1 of volume 4, chapter 2 of the IPCC guidelines for national greenhouse gas inventories (IPCC). Including the provided uncertainty, it corresponds to an initial stock of carbon varying between 50.6 and 67.2 tC/ha for high or low-activity clay soils. The corresponding mineralisation coefficient computed by Albers is 0.1176 per year. This mineralisation coefficient depends only on i) soil mean temperature and ii) clay and $CaCO_3$ content. It does not depend on site-specific parameters.

The annual input of organic carbon (m_i) comes from multiples sources:

- Exogenous inputs. Organic fertilisers (cattle manure and slurry, poultry manure and droppings, swine manure and slurry and others). National French averages of organic fertilisers use by crops (including maize and miscanthus) are listed in Table 11 of Albers's PhD thesis (Ariane Albers 2019).
- Biomass inputs incorporated into the soil after harvest. Biomass is fractioned in four carbon pools as in Albers (Ariane Albers 2019):
    - F_p (unitless, $kg_C/kg_{C,\text{ total}}$): fraction of carbon stored in the agricultural product (primary economic value), not incorporated into the soil after harvest,
    - F_s: fraction of carbon stored in the residual aboveground fraction, incorporated in the soil after harvest (residues such as straw),
    - F_r: fraction of carbon stored in root tissue (rhizome, easily recoverable), mostly incorporated in the soil after harvest if annual crops,
    - F_e: fraction of carbon stored in extra-root matter (rhizome deposition, not easily recoverable), mostly incorporated in the soil after harvest if annual crops.

The mass of carbon stored in the agricultural product ($m_{c,p}$, kg_C) can be calculated by multiplying the mass of the agricultural product by its carbon content. The relative plant carbon allocation coefficient (F_p, F_s, F_r, F_e) are given by crops (including maize and miscanthus) in Table 9 of Albers's PhD thesis (Ariane Albers 2019). These coefficients are used to calculate the mass of carbon stored in the other fractions ($m_{c,i}$, $i = s, r \text{ } or \text{ } e$) using the following formula:

$$m_{c,i} = F_i \frac{m_{c,p}}{F_p} \tag{4}$$

In the case of perennial plants, the mass of root and extra-root matter calculated correspond to an increase per year of the total mass of root and extra-root matter in the soil (Bolinder et al. 2007). A part of the roots dies each year (root senescence), is incorporated in the soil and replaced by new roots (Ledo et al. 2018). The living mass of root and extra-root matter is considered as an input to SOC only at the EoL of the plot, if the plot is ploughed for a new use (Ariane Albers 2019).

Table 14: Parameters used in the calculation of SOC changes due to the cultivation of miscanthus on marginal land in France. AG: above ground. DM: dry matter. C: carbon.

	Parameter	Value	Unit	Source
	Mean yield	6,4	tDM/ha	(Jury et al. 2022)
Yield evolution	Second year	67	% mean yield	(Colla et al. 2023)
	Third year	93	% mean yield	
	Fourth-year	100	% mean yield	
	Root senescence per year	0,17	mass dead roots/mass living roots	(Ledo et al. 2018)
Carbon partitioning	Carbon content	0,475	kgC/kg$_{\text{, total}}$	(Ariane Albers 2019)
	Agricultural product, F_p	0,268	kgC/kgC$_{\text{, total}}$	
	Residual AG fraction, F_s	0,303	kgC/kgC$_{\text{, total}}$	
	Root tissue, F_r	0,322	kgC/kgC$_{\text{, total}}$	
	Extra-root matter, F_e	0,107	kgC/kgC$_{\text{, total}}$	
Parameters for AMG	Initial total SOC stock	50-70	tC/ha	(IPCC)
	Initial stable fraction	65	% of initial total stock	(Clivot et al. 2019)
		96	% of initial total stock	(Ariane Albers 2019)
	k, mineralisation constant	0,1176	/yr	
	h, isohumic coefficient	0,126		

The IPCC guidelines for national greenhouse gas inventories (IPCC) provide default values to calculate SOC changes (volume 4, chapter 5). The initial management system is assumed to be a grassland, severely damaged (land use factor = 1, management factor = 0.7, C input level = 1). The final management system is assumed to be an idle cropland revegetated with perennial grasses, with no-till and crop residues returned to the field (land use factor = 0.93, management factor = 1.03, C input level = 1). Depending on the chosen initial carbon stock, the stock variation over the miscanthus production period (by default 20 years) is between 13 and 17 tC/ha. The inventory data to model miscanthus is taken from the work of Jury *et al.* (Jury et al. 2022). They modelled a SOC stock increase of 917 kg/ha/yr based on a presentation from Ferchaud *et al.* (Ferchaud et al. 2020), for a yield of 6.9 tDM/yr. Dufossé *et al.* (Dufossé et al. 2014) measured an increase of SOC of 13 tC/ha over the miscanthus cultivation period (20 yr) for a yield of 14.2 tDM/ha. They indicate that it is the "same order of magnitude as previous studies on this crop", varying between 6.5 tC/ha for 15 years of cultivation and 14 tC/ha for 16 years of cultivation. Hamelin *et al.* (Hamelin et al. 2012) calculated SOC variation due to miscanthus cultivation using C-TOOL. Their results vary between -0.033 tC/ha/yr for autumn miscanthus cultivated on sandy loam soil under a dry climate with a yield of 12,96 tDM/ha/yr and 0.609 tC/ha/yr for spring miscanthus produced on sandy soil under wet climate with a yield of 10 tDM/ha/yr. The values from the literature are summarised in Figure 32 and compared to the values obtained with AMG. For a static evaluation, the variation of SOC stock over the entire miscanthus cultivation (15 years) is allocated equally between each harvest.

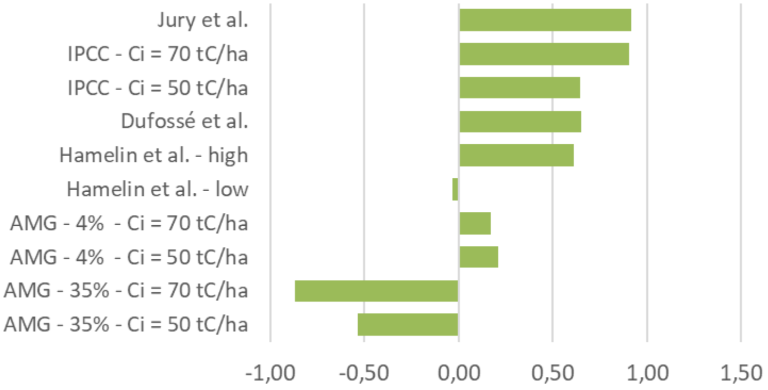

Figure 32: Calculated SOC variation with AMG compared to value from the literature. In tC/ha/yr. A negative value corresponds to an emission of CO_2.

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