Life Cycle Assessment of Two Alkaline Pretreatments of Sorghum and Miscanthus and of Their Batch Co-digestion with Cow Manure

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Life cycle assessment of two alkaline pretreatments of sorghum and miscanthus and of their batch co-digestion with cow manure.

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Abstract

In the context of bioenergy production, sorghum and miscanthus are relevant candidates for biogas production. As for many lignocellulosic biomasses, pretreatments can improve the accessibility of carbohydrates for microorganisms during anaerobic digestion. The objectives of this study were: (1) to assess environmental impacts of lime and soda pretreatments of both biomasses in codigestion with manure and (2) to compare the heat produced from natural gas with heat produced from biomethane generated from whole plants of sorghum and miscanthus under different studied scenarios. A comprehensive attributional life cycle analysis (LCA) was performed on 21 sorghum scenarios and 12 miscanthus scenarios. Certain scenarios explored direct and indirect Land Use Change (dLUC and iLUC). An environmental evaluation highlighted that most of the impacts are generated by crop production and by the purification and injection step for both sorghum and miscanthus. Compared to natural gas, the study emphasized that, unlike lime treatment, soda treatment does not provide an added value. Although most impacts are favourable towards natural gas, sorghum-based methane presents very good results (below 0) for six impact categories. The reduction of climate change ranges from -90% to -105%. Miscanthus can reduce climate change by -60% to -80%, but almost all other impact categories are in favour of natural gas. Lime pretreatment always presents best results. For both sorghum and miscanthus, it is crucial that crops are not cultivated on a land in competition with food production.

Keywords: lignocellulosic biomass; cow manure; anaerobic digestion; alkaline pretreatment; LCA; Climate change

Introduction

Since 20 years, fossil energy sources have gradually been decreasing, thus inducing the development of alternative renewable energy and the emergence of the biorefinery concept for the production of bio-based materials, chemicals and energies [1]. Renewable energies from biomass are presently widely investigated and developed [2]. In particular, biogas production by anaerobic digestion (AD) of waste and agricultural, industrial and municipal residues has become one of the most attractive renewable energy pathways [3] [4]. Biogas is valorized as electricity or heat, but for new, upgraded biogas plants, it is being increasingly injected into the natural gas network [5]. Biogas upgrading and biomethane injection into the natural gas grid has strongly risen in France due to feed-in-tariffs introduced in 2014. This alternative route towards power and heat production by combined heat and power (CHP) results in greater energy utilization yields, in particular in rural areas where heat energy is not entirely exploited. It also allows for biomethane to be stored in the gas grid [5]. Digestates are mainly valorized as organic fertilizer, while the production of high value products is still under investigation [6, 7]. This sector has faced a strong development in Europe since the 2000's, mainly in Germany and Italy, which is dominated by a model based on energy crops, such as maize, (around 35-38%), followed by manure (around 33% of the net primary energy produced from the EU biogas)[5]. However, the competition between energy crops and food and feed crops remains an important issue [7]. AD of manure alone results in low methane generation due to moderate anaerobic biodegradability of about 45–50 % [8]. This highlights the relevance of manure codigestion with energy crops or crop residues. Codigestion of manure and lignocellulosic biomass has the advantage of steadying the C/N ratio; indeed, manure presents a high nitrogen content whereas lignocellulosic biomass has a high carbon content. Moreover, manure adds a buffer capacity to the mixture. Cuetos *et al.* (2011) studied the co-digestion of swine manure with different energy crop residues (maize, rapeseed or sunflower residues) [9]. Results were different according to the type of biomass residue and a specific improvement in gas production only occurred in batch conditions, and not during semi-continuous ones. Energy crops with higher methane potentials may be more adapted for codigestion with manure, however non-food energy crops should thus be explored as alternative [10]. Koçar et al. (2013), highlighted the relevance of using C4 crops that are resistant to aridity, and have a high photosynthetic yield and CO_2 capture rate in comparison with C3 crops, thus producing more biomass [7]. In this context, the Biomass for the Future project (BFF https://www6.inrae.fr/biomassforthefuture_eng/) focused on two C4 crops, sorghum and miscanthus, and aimed at developing chain values, including anaerobic digestion, with a limited environmental footprint.

Sorghum was one of energy crops that has been highlighted for biorefineries due to a high biomass yield potential, ease of culture and wide adaptability [11]. Indeed, sorghum is cultivated under low-input agro-systems and it is particularly adapted to temperate climates. Biomass sorghum or forage sorghum is characterized by a high biomass potential [12]. This can be greater than 30 t ha^{-1} y^{-1} (reported under temperate climates) [13]. Other advantages include the excellent nitrogen usage efficiency, drought resistance, lodging tolerance and salinity resistance [14] [15]. It can be grown either as dedicated or catch crops (in double cropping systems).

Miscanthus is a promising feedstock for bioenergy production since it is a perennial crop combining high biomass production with low environmental impacts [16]. Moreover, it can grow in polluted [17] or marginal soils [18]. In particular, it can be cultivated on low quality soils with no required fertilisation [18]. In addition, the cultivation of this efficient nutrient-recycling crop can reduce nutrient run-off and increase soil organic carbon [19]. Studies have demonstrated that early-harvested miscanthus (in autumn, before senescence) has a higher biomethane potential than late-harvested miscanthus (in winter, after senescence) [20, 21]. However, early harvesting tends to reduce biomass yields later in the following years, remove nitrogen from the plant and reduce remobilization of other nutrients and carbohydrates [22]. Focus is therefore put on late-harvested miscanthus in the present study.

These types of biomass are characterized by their cellulose and lignin content [23]. Lignocellulosic biomass is often pretreated before AD because of its recalcitrance to biological conversions. Indeed, its compact structure and the bonds between polysaccharides and non-biodegradable lignin make it difficult to break down [24, 25]. Among the different kinds of pretreatment which have been widely investigated (biological, physical and chemical)[26], alkaline pretreatments have proved to be most efficient in lignin removal [27]. Moreover, both manure and crops represent solid feedstocks for which solid-state AD (SS-AD) is adapted. This process, performed with a high total solid content is characterized by a high biogas production per volume of reactor but also by a low biogas yield. Furthermore, it also presents potential instability when fed with lignocellulosic biomass, hence the relevance of pretreatments [28]. A process with growing importance in the development of the agricultural AD sector is the leach bed reactor (LBR). Here, the solid substrate is loaded into the reactor while a liquid phase, is regularly sprinkled over the solid bulk and percolates through it.

From the point of view of an environmental evaluation, covering electricity production from biogas in the Po Valley, Italy, Agostini and al. (2016) reported that codigestion of manure with a maximum of 30% sorghum entailed both greenhouse gas (GHG) emission savings and economic profit [29]. Furthermore, sorghum was found to be more profitable to cultivate than maize. In contrast, the AD of silage maize or sorghum alone provided no or very limited reduction in the climate change impact and, according to the current feed-in tariffs, generated economic losses [29]. These results were confirmed by Tsapekos et al., (2019) who reported that the mono-digestion of grass with a specific biomethane yield of 329 mL CH_4 g^{-1} organic matter (OM) would not guarantee a long-term sustainable energy system while codigestion of grass with manure ensures a sustanaible energy system, particularly under reduced transportation scenarios with an average distance between the grasslands and biogas plant of less than 50 km [30].

These studies have thus confirmed that manure digestion would be the most efficient way to reduce GHG emissions, although trade-offs do exist with other local environmental impacts. Indeed, the critical points in biogas production processes involve different variables at regional scale studies, such as climate, raw material availability, transportation distance, source of inputs and energy, plant location and technological development level [31]. According to O'Keeffe et al. (2020), AD represents a significant decentralized renewable energy technology for mitigating climate change [32]. However, it depends on local and regional feedstocks, which determine its sustainability [32]. Due to its low energy value and low biogas conversion efficiency, manure is indicated for use in co-digestion with other substrates to produce biogas. Its application to biogas production would not only be a profitable way to reduce impacts on the environment, but also to produce energy and biofertilizers.

On the basis of the review of 15 LCA studies on biogas systems across Europe, Hijazi et al. (2016) confirmed that the type of feedstock is a crucial parameter in the environmental impact of biogas systems [33]. In the case where crops are cultivated for biogas production, the environmental impacts from their production can become dominant within the biogas system. More particularly Gonzales-Garcia et al. (2013) demonstrated how mineral fertilisation leads to the highest contribution in biogas systems from maize, wheat or triticale [34]. For sorghum and miscanthus, it is thus important to analyse the different possible scenarios for their cropping. In addition, by employing two different grass harvesting machines, Tsapekos et al. (2018) highlighted the impact of mechanical pretreatments on environmental impacts [35]. However, to our best knowledge, no study has yet considered alkali pretreatments in LCA analysis of biogas systems.

The objectives of this paper are (1) to examine the environmental impacts of alkaline pretreatments of sorghum and miscanthus when they are co-digested with cattle manure and (2) to compare heat produced from biomethane generated from both biomasses in the different studied scenarios with heat produced from natural gas. The studied AD systems are batch leach bed reactors. Performance of alkaline pretreatments and codigestion were investigated at lab-scales. For sorghum they are presented in the supplementary material (table S1) while for miscanthus, the data originate from a previous study [36].

2.Material and methods

2.1 Description of scenarios

This study focusses on the methane production from 15% sorghum (whole plant except for the lower part of stem that remains on the ground; 30% Dry Matter (DM)) or early spring harvested miscanthus (85% DM) with 85% cattle manure. The considered solid state anaerobic digestion process is a batch leach bed reactor with 5 feeding scenarios:

- Sorgh_Baseline: codigestion of sorghum and cattle manure with 15% (wet weight) of sorghum. This percentage is the limit authorized in France for energy crops.
- Sorgh_CaO: codigestion of 15% sorghum and cattle manure, sorghum was pretreated with 10 g lime (CaO) 100 g^{-1} DM for 5 days at ambient temperature
- Sorgh_NaOH: codigestion of 15% sorghum and cattle manure, sorghum was pretreated with 10 g soda (NaOH) 100g^{-1} DM for 1 day at ambient temperature
- Misc_Baseline: codigestion of 15% miscanthus and cattle manure
- Misc_CaO : codigestion of 15% miscanthus and cattle manure, miscanthus was pretreated with 10 g CaO 100 g^{-1} DM for 5 days at ambient temperature

The impact of pretreatments on biochemical methane potentials (BMP) and methane yields observed in the different scenarios are detailed in supplementary materials for sorghum

(Table S2) and in Thomas et al. (2018) for miscanthus [36]. Regarding the increase of methane production due to alkali pretreatments, a few discrepancies were observed between BMP and codigestion results. In order to avoid a too conservative or optimistic estimation, the average increases were used. Table 1 reports the lab-scale methane yields used in each scenario. Considering that methane production is lower in upscaled digesters, the methane yields were weighted by a 82.5% factor. In the present study a digester has been considered to be able to process 5 400 t yr^{-1}.

Table 1: Methane yields of sorghum and miscanthus and increase following alkaline pretreatment with lime (CaO) or soda (NaOH).

Scenario	Methane yield (NmL gDM^{-1})	Increase due to pretreatment
Sorgh_Baseline	250	
Sorgh_CaO	265	6 %
Sorgh_NaOH	283	13 %
Misc_Baseline	158	
Misc_CaO	193	22 %

Different sub-scenarios which were considered for sorghum and miscanthus production are described in section 2.2.

2.2 Functional unit and reference flows

The present study focusses on the production of industrial heat from sorghum or miscanthus-produced methane during co-digestion with cattle manure, which has been injected into the gas grid, and its comparison with the production of industrial heat from natural gas based on a typical French context.

The function of the system is the production of industrial heat. The functional unit, the baseline comparison between scenarios, and the reference flows for each scenario are reported in Table 2. The different scenarios are representative of co-digestion without pretreatment or with a CaO or NaOH treatment. It is noteworthy that the average production of raw materials, especially crops, have been taken into account. This is particularly important for miscanthus since its average production is determined over 15 years. Indeed, there is no production the first year, a low production the second and third years, and full production is only reached between years four and fifteen.

Table 2: Function, functional units and reference flow for each scenario

Function	Scenario	Functional unit	Reference flow
Heat production from miscanthus	Misc_Baseline	1 MJ	$2.64\ 10^{-1}$ kg Dry Matter (DM)
	Misc_CaO	1 MJ	$2.04\ 10^{-1}$ kg DM
Heat production from sorghum	Sorgh_Baseline	1 MJ	$2.00\ 10^{-1}$ kg DM
	Sorgh_CaO	1 MJ	$1.75\ 10^{-1}$ kg DM
	Sorgh_NaOH		$1.49\ 10^{-1}$ kg DM
Heat production from natural gas	Natural gas	1 MJ	$2.51\ 10^{-1}$ Nm^3

The production of heat from codigested sorghum or miscanthus derived methane are not expected to have large-scale consequences on any markets. The LCA was therefore performed according to the methodology recommended by the European Union in the frame of the International Life Cycle Database (ILCD) for a "Micro-level decision support" LCA's type [37]. In summary, this implies that (i) only direct consequences are to be taken into account, (ii) background processes can be determined in an attributional manner and (iii) the co-function that cannot be solved by a subdivision of the system ought to be be solved in priority by the system expansion approach and, finally by an allocation.

The co-digestion of sorghum or miscanthus with cattle manure provides two main functions (1) cattle manure treatment and (2) biogas production. However, the valorisation of the anaerobic digestion waste, *i.e.* the digestate, allows for mineral fertiliser production, transport, spreading and on-field emissions to be avoided. This co-function is first solved by allocation of the digestate to each main function and secondly, by the system expansion approach. Table 3 reports the methodologies applied to solve the multi-functionalities of co-digestion in order to build foreground data for sorghum and miscanthus. This table also reports the methodology used for solving the co-function linked to the spreading of digestate as a fertiliser.

Table 3: Methodologies applied to solve multi-functionalities

Flow	Methodologies applied and associated share
Biomass production	Sub-division: 100% associated to sorghum or miscanthus scenarios; 0% to cattle manure treatment
Pretreatment	Sub-division: 100% associated to sorghum or miscanthus scenarios; 0% to cattle manure treatment
Anaerobic digestion inputs	Mass allocation: all the anaerobic digestion consumptions (fuel, electricity and heat) are presumed to be related to the amount of input material handled; 15% for miscanthus or sorghum; 85% for cattle manure treatment
Digestate handling	Mass allocation: the amount of digestate produced is presumed to be directly related to the input mass; Thus, 15% of digestate storage, transport and spreading is allocated to miscanthus or sorghum; 85% to cattle manure treatment
Digestate composition	Sub-division: The NPK composition of digestate, required for the determination of storage and on -field spreading emissions as well as the agricultural value are defined according to the miscanthus or sorghum composition
Digestate agricultural value	System expansion: The co-function held by the spreading and valorisation of digestate nutrients is solved according to the avoided production, transport, spreading and on-field emissions of an equal amount of useful N, P and K mineral fertilisers

It is noteworthy that although LCA should only consider the direct consequences of the system, the influence of indirect consequences due to Land Use Change (iLUC) have been investigated.

Although iLUC occurs when biomass use presents a large-scale influence on the food market, this should not be the case in the studied scenarios. However, some authors argue that any soil use contributes to the global use of soil and thus, among others, to tropical deforestation [38–40]. Since it has been proved that such iLUC may counterbalance the advantages of biobased energy, this paper aims at investigating, in a simple manner, the risk of iLUC of producing heat from sorghum or miscanthus biomethane [41].

2.3 System boundaries and sub-scenarios

This is a cradle-to-grave LCA. It considers the main processes, resource consumption and waste from the production of miscanthus to the combustion of the biomethane injected into the natural gas grid according to French conditions, as reported in Figure 1-a and in Figure 1-b for natural gas.

Sub-scenarios were considered for miscanthus production. Two different yields of dry matter (DM) and agricultural practices have been determined by BFF project partners. These involve a highly productive land with a 14.8 t DM ha^{-1} $year^{-1}$ (HighProdLand or HPL) and a lower productive land, with 6.4 t DM ha^{-1} $year^{-1}$ (LowProdLand or LPL), that requires fertilisation.

As reported in table 4, no direct neither indirect consequences of Land Use Change (dLUC/iLUC) are considered in the baseline scenario. It represents a scenario where miscanthus has been cultivated for over 20 years on the same field which is not in competition with food production (marginal land). This scenario is presumed to be the best representation of the performance of miscanthus over a mid-term perspective, once the sector has been developed at an industrial scale and the dLUC and iLUC are not relevant anymore. Two additional scenarios were modelled. They represent the performances for the first 20 years of the miscanthus cultivation, before steady state is reached between the direct and indirect variations in Soil Organic Carbon (SOC) stock. In the first scenario, called dLUC, miscanthus is cultivated on a marginal land that is not in competition with food production and is characterised by a high SOC. In this case cultivation leads to a SOC variation from -0.5 to 0 t C ha^{-1} y^{-1} over 20 years [Ferchaud 2020] and consequently to CO_2 emissions. An average value of -0.25 t C ha^{-1} y^{-1} has been used. The second scenario, called d+iLUC, is representative of miscanthus cultivation on land where there is competition with food production. In this case the miscanthus grows in a soil characterised by a low SOC stock and leads to a direct SOC variation from +0.2 to +0.6 t C ha^{-1} y^{-1} over 20 years, thus implying CO_2 storage[42]. An average value of -0.4 t C ha^{-1} y^{-1} has been used. Finally, considering that the demand on crops for non-food application is strongly increasing, the influence of possible indirect consequences due to LUC has been explored. According to Audsley et al. (2009), Schmidt et al. (2011) and Flysjö et al. (2012), the indirect CO_2 emissions range from 1.43 to 8.58 t CO_2 ha^{-1} y^{-1}[38][39][43]. An average value of 5.00 t CO_2 ha^{-1} $y^{-1}$was used (1.36 t C ha^{-1} y^{-1}). Moreover, in a rough estimation, the use of 1 ha to produce miscanthus in France instead of food crops has been presumed to entail the transformation and occupation of 1 ha of tropical forest.

Table 4: Summary of direct and indirect Soil Organic Carbon (SOC) variation according to each sub-scenario*

Cultivation	dLUC	iLUC	Unit
Over 20 years, Marginal	0	0	
Less than 20 years; Marginal	-917	0	kg CO_2 ha^{-1} y-^{1}
Less than 20 years; Non-Marginal	1467	-5000	kg CO_2 ha^{-1} y^{-1}

* A negative variation of SOC leads to CO_2 emissions and a positive variation leads to CO_2 storage in soil.

Five sub scenarios were considered for the production of sorghum (Table 5). They were set up by the different partners of the BFF project. In three scenarios, sorghum is cultivated in majority on the field and in two scenarios it is cultivated as a catch crop. Crop yields range from 7 to 14.5 t DM ha^{-1}. Different agricultural practices (irrigation, fertilisation, roots depth, seeding) were investigated according to the region (south/north of France) and to the soil depth. In all cases, presumption was made that the field had been cultivated for more than 20 years and that sorghum cultivation does not modify the soil carbon stock (no dLUC). The consequences of iLUC was assessed during the sensitivity analysis where sorghum was considered to be the main culture on the field. The same approach was applied for miscanthus.

Table 5: Definition of the sorghum cultivation sub-scenarios

Scenario	Type of culture	Soil depth	France region	Yield (t DM ha^{-1})
Main_Int_North	Main	Intermediate	North	10
Main_Int_South	Main	Intermediate	South	14.5
Main_Shal_South	Main	Shallow	South	12
Catch_Int_South	Catch	Intermediate	South	10
Catch_Shal_South	Catch	Shallow	South	7

2.4 Life cycle inventory and result calculations

The assessment was performed on the LCA software GaBi®. Table 6 reports the origin of the foreground and background data to calculate the life cycle inventories (LCI). A full LCI with a scenario based on miscanthus and sorghum is reported in SI (tables S3 to S13). It reports the origin of the data with more details. The input and output of co-digestion were split between the different substrates entering co-digestion according to different rules as described in table 3. The potential environmental impacts were calculated with version 1.09 of the environmental impact evaluation set recommended by the European union in the frame of the ILCD [44]. A single score was calculated using the ReCiPe HA v1.08 method. In both cases, the carbon emissions resulting from crops were considered as biogenic. Hence, there is no impact for carbon dioxide and a reduced impact is taken into account for methane emissions.

Table 6: Origin of the foreground and background data for all scenarios. More details are given in SI (tables S3 to S13)

Steps	Foreground	Background
Crop production	- BFF partners for the miscanthus and sorghum production and transport - Agribalyse methodology for the calculation of pesticide emissions as well as nitrogen emissions due to fertilisation or crop residues - Literature for the variation of soil organic carbon	Agribalyse 1.2 database for agriculture when possible. Otherwise Ecoinvent 3.3.
Methane production, injection and transport	- BFF partners for the methane yields of miscanthus and sorghum as well as for co-digestion operations - Ecoinvent 3.3 for the purification by pressure swing absorption, injection & transport - Ecoinvent 3.3 for the alternative	Ecoinvent 3.3 (adapted as reported in SM)
Storage and spreading of digestate	- The digestate composition is based on sorghum and miscanthus composition and degradation yields during anaerobic digestion - Assumption for transport distance and spreading - IPCC (2006a) for the calculation of nitrogen emissions due to storage and spreading - IPCC (2006b) for the calculation of SOC increase due to digestate spreading	Ecoinvent 3.3
Heat production	- Natural gas combustion from Ecoinvent3.3 adapted to the nature of the carbon for the BFF scenarios - Ecoinvent 3.3 for the alternative	Ecoinvent 3.3

3. Results

3.1 Sorghum

3.1.1 Contribution analysis

In order to simplify the interpretation of the contribution analysis, neither the LUC nor the avoided impact have been taken into account. Moreover, there is no carbon dioxide storage by crops or any emission of carbon dioxide when bio-based methane is burnt. Figure 2-a reports the contribution analysis for sorghum cultivated as an intermediary crop in the south of France on an intermediary soil and without sorghum pretreatment. The conclusions for the other cultivation scenarios are similar by a few percent up or down. There are three phases which generate the impacts. Production has a significant influence on almost all impact categories (35% on average). This result is comparable to those obtained by Boulamanti et al. (2013) who reported that maize production accounts for 28-42% of the GHG emissions of the whole process when anaerobic digestion of maize and biogas is converted to electricity [45]. This result was similarly reported for anaerobic digestion of sorghum, maize or wheat either diluted with water or codigested with manure across various regions of Italy [46]. It is noteworthy that sorghum provided the best environmental performance, followed by maize and wheat. The crop cultivation phase was the major contributor to all indicators in all scenarios, followed by electricity consumption for the plant operation [46]. Cultivation impacts of biomass production are mainly related to the production of nitrogen fertiliser (as already reported in the case of maize, wheat and triticale [34]) as well as to associated nitrogen emissions in the field (ammoniac (NH_3), nitrogen oxides (NOx), nitrate (NO_3), nitrous oxide (N_2O)). For sorghum, maize or wheat cultivation in Italy, highest cultivation impacts are also due to fertilisation, followed by ploughing and irrigation [46]. Land use is driven by the occupation of land for sorghum production. Irrigation has a significant contribution to carcinogenic human toxicity (45% of the production share), freshwater eutrophication (35% of the production share), non-cancer human toxicity (20% of the production share), freshwater ecotoxicity (20% of the production share) and water depletion (15% of the production share). The purification, injection and transport of the biomethane contribute to 25% on average. This is mainly driven by the infrastructure for the ecotoxicity and eutrophication of freshwater, as well as the cancer and non-cancer human toxicities and the depletion of mineral, fossil and renewable resources. Electricity is the main source of ionizing radiation as well as of ozone depletion because of the high percentage of nuclear power in the French electricity mix. Methane losses drive the climate change impact of purification/injection and contribute, with electricity and infrastructure, to the formation of particulate matter. However, among the technologies used for biomethane upgrading (high pressure water scrubbing, alkaline upgrading, pressure swing adsorption, membrane separation, cryogenic separation), the lowest environmental impacts were achieved by high pressure water scrubbing, followed by pressure swing adsorption which is considered in the present study [30].

The third step contributing to environmental impacts is digestate spreading. Its contribution is significant (more than 40%) for six impact categories where nitrogen emissions occurring in the field (NH_3, NOx, N_2O, NO_3) play a role: acidification, climate change, marine and terrestrial eutrophication, particulate and photochemical ozone formation. Digestate management is an important topic and several digestate management options have been proposed for environmental improvement. For example, installing gas-tight tanks for digestate storage is highly recommended [29]. According to Rehl et al. (2011), digestate solar drying or composting were suitable solutions for reducing environmental impacts [47].

The contribution analysis for the scenarios with lime pretreatment is very similar to the one without pretreatment (+/- 1%; Figure 2-b). On the contrary, the contribution analysis for the sodium hydroxide pretreatment scenario leads to various contributions (Figure 2-c). The contribution of pretreatment rises by 25%, on average. The influence of NaOH production is especially relevant for ozone depletion (70% of the overall impact), water depletion (50% of the overall impact), eutrophication and ecotoxicity of freshwater (45% and 40% of the overall impact), human toxicities (35% and 20% of the overall impact) as well as mineral, fossil and renewable resource depletion, particulate matter formation and climate change (more or less 20% of each of these categories). The increase of these contributions leads to a fall in the production contribution by 10% and of the purification/injection and digestate spreading influence by 5% each. In a study where different pretreatment techniques for second generation bioethanol production were compared, the environmental impacts of alkali pretreatment with sodium hydroxide were found to be worse than with thermal pretreatments (steam explosion or hot water pretreatment) but better than dilute acid pretreatments [48].

Figure 3-a illustrates the influence of avoided impacts due to the fertilisation and increase of carbon soil stock related to digestate for sorghum cultivated as an intermediary crop in the south of France on an intermediary soil and without sorghum pretreatment. This influence is significant (-50% at least and -85% on average) for all impact categories, excepted land use (-10%), because of the avoided production of mineral nitrogen fertiliser and nitrogen emissions (NOx, NH_3, NO_3, N_2O).This result concurs with results from Lijó et al. (2014) for codigestion of manure with maize and triticale silages in Italy and from de Meester et al. (2012) where digestate was used for maize cultivation in Germany [49, 50]. In contrast, avoided emissions due to the use of digestate as fertiliser was not found to be very significant for anaerobic digestion of sorghum, maize or wheat either diluted with water or codigested with manure [46]. According to Montemayor et al. (2019), the replacement of a large portion of mineral fertilisers with digestate could offset the impact of all freshwater eutrophication, ozone depletion by 96-99%, resource depletion by 94-96%, global warming by 25-35 %, photochemical ozone formation by 17-22 but it greatly contributes to acidification (51%) [51]. The influence of avoided impacts is slightly lower in the lime pretreatment scenario (-80% on average instead of -85%; Fig 3-b) and much lower for the sodium hydroxide pretreatment (-50% on average instead of -85%; Fig. 3-c). This is due to the fact that the avoided impact of digestate spreading does not vary while the life cycle impact increases slightly for the lime pretreatment and strongly for the sodium hydroxide pretreatment. Nevertheless, note that at this level, the benefit related to the increase of methane and heat production is not taken into account. This ought to be the case when heat production from sorghum-based methane is compared with heat from natural gas.

3.1.2 Comparison with natural gas

The comparison of the natural gas with the five scenarios of sorghum cultivation without pretreatment is illustrated in Figure 4. The results from sorghum are normalized to those of the natural gas. Hence, if the results from sorghum are better than those of natural gas, they should be below 100% and vice versa. Figure 4-a depicts how nine categories out of fifteen are undoubtedly in favour of natural gas (sorghum > by a factor 13 at least). This figure points out that there is scarcely any difference between the modelled scenarios except for marine eutrophication or when iLUC is taken into account. Figure 4-b, reports that about six impact categories out of fifteen are in favour of sorghum. Four of them report results below zero, thus implying the benefit is greater than generated impact. This could also concern the two remaining impact categories, although they depend on the scenarios. As for water depletion, the scenarios with an irrigation ratio above 80 m^3 t^{-1} DM present a worse impact than for natural gas. Indeed, when the ratio is less than 60 m^3 t^{-1} DM, the benefit is greater than the impact. All scenarios show a strong reduction in climate change (-87% to -105%) although the risk of iLUC for the scenario where sorghum represents the major crop in the field counterbalances the benefit.

According to Figure 5, certain impact categories are improved (-10% to -2200%) and others are penalised (+5% to +700%) by the lime pretreatment in comparison to no pretreatment. Nevertheless, the ranking with natural gas is not modified. The only exception observed is for water depletion: indeed, no scenario is favourable towards heat produced from sorghum-based methane. Another noteworthy observation is that climate change is still largely promising with sorghum (-8% to -100%) if iLUC is not considered. If iLUC should be taken into account the benefit on climate change is once more counterbalanced although sorghum-based methane still presents better results (-16%) for the scenario where sorghum is the main cultivated crop in an intermediate soil in the south of France.

Although certain impact categories are favourable to sodium hydroxide pretreatment (-700% to -3400% in comparison with no pretreatment, Figure 6), the impact does not turn in favour of sorghum. On the contrary, many impact categories can be penalised by sodium hydroxide treatment (30% to 6000%) and many turn in favour of natural gas and especially climate change.

The detailed results for all scenarios regarding climate change are available in table 7. Results range from – 3.6 10^{-3} kg CO_2eq MJ^{-1} (main Int South, no pretreatment, no iLUC) to + 9.86 10^{-2} kg CO_2eq MJ^{-1} (main Int North, no pretreatment, iLUC). An improved global warming impact was obtained in the case of co-digestion of manure with sorghum grown in Italy : -0.104 kg CO_2eq MJ^{-1}[46].

Table 7: Climate change impact for each sorghum scenario and natural gas

Pretreatment	Scenario name[a]	Results (kg CO_2eq J^{-1})	
		NoLUC	iLUC
No	Main_Int_North	-1.12E-03	9.86E-02
	Main_Int_South	-3.60E-03	6.50E-02
	Main_Shal_South	6.22E-03	8.94E-02
	Catch_Int_South	1.21E-03	-
	Catch_Shal_South	8.97E-03	-
CaO	Main_Int_North	2.41E-03	8.95E-02
	Main_Int_South	-1.00E-04	5.98E-02
	Main_Shal_South	8.82E-03	8.13E-02
	Catch_Int_South	4.45E-03	-
	Catch_Shal_South	1.12E-02	-
NaOH	Main_Int_North	2.41E-02	9.82E-02
	Main_Int_South	2.20E-02	7.29E-02
	Main_Shal_South	2.96E-02	9.12E-02
	Catch_Int_South	2.58E-02	-
	Catch_Shal_South	3.16E-02	-
No	Natural gas	7.13E-02	-

[a] See Table 5 for the definition of the Sub-scenarios,
CaO: lime; NaOH : soda
dLUC: direct Land Use Change; iLUC: indirect Land Use Change

The comparison was also performed using the ReCiPe HA (Europe) method in order to verify whether the conclusions were similar to those obtained through the midpoint approach. When the cultivation of sorghum is considered to be the main crop on a plot of land, thus with the risk of iLUC, the single score is driven by land transformation in tropical areas (more than 80%) and is much higher than for natural gas (+650% to 950%, see Figure S1 in supplementary information). Figure 7-a reports the single score for scenarios without treatment. If the impacts that are avoided thanks to digestate application are taken into account, all scenarios present a better single score (-15% to - 40%) in comparison with the production of heat using natural gas except when sorghum is cultivated as an intermediate low productivity (7 t DM ha^{-1}) crop. Figure 7-b presents the single score in case of lime pretreatment. Results are very similar to cases without pretreatment but it is noteworthy that although it increases the impact on the climate change, the decrease in particulate matter formation and agricultural land occupation tends to improve the single score by -3% to -10%. Finally, as expected from the midpoint analysis, the sodium hydroxide pretreatment increases the single score in comparison with cases without pretreatment (from 10% to 25%). Hence, from a global point of view, only the scenario where sorghum is cultivated as the main crop in an intermediary soil in the south of France proves to be competitive (Figure 7-c). The decrease of the single score by the avoided impact of digestate is due to climate change (50%), fossil depletion and particulate matter production (20% each).

3.2. Miscanthus

3.2.1 Contribution analysis

Figure 8 reports the contribution analysis of the different stages from biomass production to methane combustion. For easier interpretation of the contribution analysis, no LUC, neither avoided impact due to digestate use, were taken into account. Moreover, neither carbon dioxide storage by crops nor carbon dioxide emissions appear when biomethane is burnt. For the miscanthus cultivated on highly productive land (HPL), three phases can generate impacts (Figure 8-a). Production has a strong influence on land use (because of land occupation) as well as water eutrophication due to nitrogen and phosphorus emissions from crop residues and agricultural soil losses. The purification, injection and transport of biomethane contribute largely for almost all impact categories (40% on average and up to 90%). This is essentially driven by the infrastructure for the ecotoxicity and eutrophication of freshwater, the cancer and non-cancer human toxicities as well as the depletion of mineral, fossil and renewable resources. Electricity is the main source of ionizing radiation as well as of ozone depletion because of the high share in nuclear power within the French electricity mix. Methane losses drive the climate change impact of the purification/injection and contribute with electricity and infrastructure to particulate matter formation. The third step, contributing to the environmental impact, is digestate spreading through nitrogen emissions (NH_3, NOx, N_2O, NO_3) occurring in the field.

For the miscanthus cultivated on LPL (Figure 8-b), the effects of cultivation increase strongly and become the first source of impact (45% in average over all categories). The lower yield per hectare leads to the use of more surface and machinery to obtain the same amount of miscanthus. Moreover, this scenario takes into account the use of nitrogen fertiliser that in turn contributes to increase nitrogen emissions and thus the acidification, climate change, marine and terrestrial eutrophication, particulate matter and photochemical ozone formation impacts. Krzyzaniak et al. (2020) reported that the use of fertiliser had the greatest impact on freshwater eutrophication and terrestrial acidification for miscanthus cultivation [18]. Nevertheless, according to Kisesel et al. (2016) these impacts, as well as climate change, fossil fuel depletion and marine eutrophication, could be reduced if miscanthus were used in biogas plants instead of maize [19]. The contribution analysis for the scenario including lime pretreatment is very similar to the no pretreatment scenario ((Figures 8-c and 8-d). The use of CaO is almost undetected but increases the production of methane and thus the weight of the purification/injection step by 5% at the maximum.

Figure 9-a illustrates the influence of avoided impacts due to the fertilisation and increase of carbon soil stock related to digestate for the HPL scenario. The effect is significant (-20% to -45%) on acidification, climate change, marine and terrestrial eutrophication, ozone depletion as well as particulate matter and photochemical ozone formation because of the avoided production of mineral nitrogen fertiliser and nitrogen emissions (NOx, NH_3, NO_3, N_2O). The effect is lower for the LPL scenario although it allows the previously listed impact categories to decrease by about 10% to 20% (Figure 9-b). Lime pretreatment has very few effects on the contribution analysis of heat production. In consequence, the influence of the avoided impact due to the use of digestate as fertiliser is very similar to the no pretreatment HPL and LPL scenarios (Figures 9-c and d).

3.2.2 Comparison with natural gas

Comparison with natural gas is illustrated in figure 10 for miscanthus cultivated on LPL and figure 11 for miscanthus cultivated on HPL. Whatever the considered scenario, 13 to 14 impact categories out of 15 favour natural gas. The ozone layer depletion is always in favour of the miscanthus scenarios. The comparison between climate change impacts varies strongly. When miscanthus is cultivated on an LPL (figure 10-b), the only scenarios showing better performances than for natural gas concern miscanthus that had been cultivated for more than 20 years and for which no LUC occurs (-34% without pretreatment and -42% with lime treatment). Otherwise, results are equivalent or worst, especially considering the consequences due to iLUC.

When miscanthus has been cultivated on an HPL (figure 11-b) for more than 20 years, the climate change impact is better than for natural gas (-82% without lime treatment and -79% with lime treatment). When miscanthus has been cultivated on a marginal land, and not in competition with food production, *i.e.* only the dLUC is taken into account, results are still better than for natural gas (-57% without pretreatment and -60 with pretreatment). When miscanthus has been cultivated on a land in competition with food production, the indirect CO_2 emissions due to iLUC counterbalance the benefits of miscanthus based methane (+11% without lime treatment and -8% with pretreatment).

Detailed results for all scenarios regarding climate change are available in table 8.

Global warming for miscanthus ranges from 1.30 10^{-2} kg CO_2eq MJ^{-1} (HPL, no pretreatment, no LUC) to 2.01 10^{-2} kg CO_2eq MJ^{-1} (LPL, iLUC) which corresponds to 1.215 and 8.02 t

CO_2eq ha^{-1}, respectively. In a study comparing various perennial crops (*Miscanthus x giganteus*, ryegrass and willow for bioenergy production, values were estimated between -4.1 and 13.5 t CO_2eq ha^{-1} y^{-1} [52]. The authors highlighted these high values due to iLUC which had not been considered in previous studies. For example in a bioenergy system based on miscanthus in Italy, Fazio and Monti (2011) observed values up to -25 t CO_2eq ha^{-1} y^{-1} [53]. These results have been confirmed for anaerobic digestion of miscanthus where the net global warming potential rose from -22 t CO_2eq ha^{-1} y^{-1} to +11 t CO_2eq ha^{-1} y^{-1} when iLUC was integrated in the calculation [54]. In contrast, even with no LUC, greenhouse gas emissions of 4.1 t CO_2eq ha^{-1} were reported for miscanthus grown in Poland [55].

Table 8 : Climate change impact for each miscanthus scenario and for natural gas

		Results (kg CO2eq MJ^{-1})		
Pretreatment	Scenario name	No-LUC (>20y)	dLUC (marginal <20 y)	d+iLUC (non-marginal <20 y)
No	HPL	1.30E-02	3.04E-02	7.88E-02
	LPL	4.72E-02	8.73E-02	2.01E-01
CaO	HPL	1.48E-02	2.84E-02	6.57E-02
	LPL	4.14E-02	7.24E-02	1.60E-01
No	Natural gas	7.13E-02	7.13E-02	7.13E-02

HPL: High Productive Land; LPL: Low Productive Land; dLUC: direct Land Use Change; iLUC: indirect Land Use Change

The comparison was also performed using the ReCiPe HA (Europe) method in order to verify whether the conclusions were similar to those obtained thanks to the midpoint approach. When cultivation on a plot of land in competition with food production was considered, the single score of miscanthus scenarios were driven by land transformation (more than 74%) in tropical areas and results were 800% to 2500% higher than for natural gas; results are reported in the Supplementary information (Figure S2). For the LPL, the single score analysis led to the same conclusion as the midpoint analysis (Figure 12): miscanthus does not seem to be an alternative to natural gas for heat production, even when lime pre-treatment is considered. However, if miscanthus should be cultivated on an HPL (Figure 12), the single score clearly becomes more competitive (+13%) and can be slightly better to natural gas as long as lime pretreatment is applied (-7%). In this very specific case, it appears that the production of heat from miscanthus-based methane contributes to reduce the climate change impact without increasing the overall environmental single score.

3.3 Discussion, limitations of the study

Firstly, certain methodological points in this study could be improved. In particular, the main estimation of nitrogen (NO_3, NOx, N_2O, NH_3) was based on that used in the Agribalyse database for the LCA of French crop production. Note however that this was not developed for permanent crops such as miscanthus. Improved modelling of digestate emissions and benefits according to fertilisation practices would help to improve environmental performances as well as the robustness of the analysis. The impact of biomethane purification and injection was extracted from the ecoinvent database. Therefore both the representativness of the data as well as the design of the infrastructure could still be improved to enhance the evaluation. Emissions from natural gas extraction and transport are usually underestimated in LCA studies. Indeed, Gruber and Brandt (2019) reported that methane leakages from natural gas systems are inconsistently characterized and likely systematically underestimated by commonly used life cycle inventory (LCI) databases [56]. Methane emissions can significantly vary according the system, median values are estimated 0.8-2.2% of total methane production whereas mean emission values range from 1.6 to 5.5%. The median CO_2 equivalent emissions are estimated to 18-24 g CO_2 eq MJ^{-1} and the mean ones to 22-107 g CO_2 eq MJ^{-1}, calculated on the higher heating value (HHV) [57].

Estimation of the consequences of iLUC could also be significantly improved since several approximate assumptions have been made (i.e., that 1 ha of miscanthus or sorghum cultivated in competition with food production replaces 1 ha of primary forest). The consequences of the iLUC have probably been overestimated since it is likely that 1 ha cultivated in France will not genuinely lead to the deforestation of 1 ha of tropical forest. The reduction of food production in France could indeed be partially compensated by other means than tropical deforestation such as an increase in fertilisation, the improvement of agricultural practices or intensification or a reduced consumption [58]. Finally, last but not least, the yield on deforested tropical areas is likely to be higher than the yield on an arable land in France, especially when LPL is taken into account. In the present study conditions, iLUC, when taken into account, counterbalanced the environmental benefit on climate change, except for the scenario where sorghum is the main crop cultivated on an intermediate soil in the south of France. This result was also highlighted for cases of co-digestion of different perennial crops with manure, when impacts of global warming were less favourable than the fossil gas reference and the iLUC impact represented a paramount average of 41% of induced greenhouse gas emissions [52].

It is noteworthy that the sorghum scenario with no pretreatment, leading to better impacts than natural gas, presents the highest biomass yield. As expected, this parameter has the strongest influence in similar studies. The biomass yields, which were defined by a panel of specialists in French agriculture, can be considered as reliable. However, if these values are compared to published data, they are low or within the lowest range. Indeed, sorghum yields considered in this study ranged from 7 to 10 t DM ha^{-1} and 10 to 14.5 t DM ha^{-1} for catch and main crops, respectively. For sweet sorghum, Jankowski et al.(2020) measured 17.7 t DM ha^{-1} over a 11-year field experiment in Poland and mentioned yields ranging from 10.4 to 25.5 t DM ha^{-1} in central Europe [59]. In Italy, yields of 22 [53] and 28 [46] t DM ha^{-1} were considered for LCA studies, the latter being almost the double of the highest yield considered in the present study. As for miscanthus, the different scenarios considered biomass yields from 6.4 to 14.8 t DM $ha^{-1.}$ These yields are within the lowest range of values reported in the literature. In southern Germany, according to the year and location, biomass yields of genotype *Miscanthus x giganteus* varied from 10 to 30 t DM ha^{-1} if harvested in December, and 16-18% lower yields were obtained if the harvest was delayed to February [60]. In Italy, Fazio reported a yield of 17 t DM ha^{-1} whereas it was estimated to 17.8 t DM ha^{-1} in Poland [55] and to 10 ± 3.3 t DM ha^{-1} in Denmark [52]. On the base of a review paper, average yields of *M. x giganteus* species during the build-up period were reported to be 5.9, 8.3 and 13.0 t DM ha^{-1} for the first, second and third year, respectively [16]. During the adult plateau phase, the yield is higher. For example, in the north of France, Arnoult et al. (2015) measured, during the winter, an average biomass yield of 19 *Miscanthus* clones harvested of 2.2, 8.7, 24.7, 14.5 and 17.3 t DM ha^{-1} for the 2nd, 3rd, 4th, 5th and 6th year [61]. Wagner et al. (2019) built their LCA study on 18 t DM ha^{-1}yield for miscanthus grown on marginal land. However since they stated that such yields could have been significantly lower, they performed a sensitivity analysis with yields down to 6 t DM ha^{-1}[62]. Results pointed out that with yields lower than 9 t DM ha^{-1}, the substitution of the fossil reference led to a net impact on fresh-water eutrophication, while benefits were lower in climate change, freshwater and marine ecotoxicity.

The methane yield per hectare or per amount of dry matter is also a very important factor. The value determined in this study 250 NmL g^{-1}DM is lower than values used in LCA studies (313 NmL g^{-1} DM [59] or 341 NmL g^{-1} DM [46] but is within the range of methane potentials reported in the literature (220 NmL g^{-1} DM [63],from 248 to 277 NmL g^{-1} DM for 5 sorghum varieties [64], 243 NmL g^{-1} DM for a fibre sorghum [65], from 242 to 256 NmL g^{-1} DM for 3 sorghum varieties [66], from 192 to 342 NmL g^{-1} DM over a panel of 57 sorghum genotypes [67]. As for miscanthus, the methane potential used in this study (158 NmL g^{-1} DM) was within the range of values published for *M. x giganteus* harvested in winter. However, when harvested in autumn, methane potentials are generally higher. For example, values ranging from 285 to 333 NmL g^{-1} Organic Matter (OM) were reported for *M. x giganteus* harvested between end of August and mid-november [20]. Lower methane potentials were reported in the case of *M. x giganteus* grown in Denmark: 159, 110 and 98 NmL g^{-1} OM when it was harvested in September, February and April, respectively [68]. An early harvest in autumn implies a higher methane potential (+36%) than for a harvest in February as considered in the present study. However an autumn harvest would not allow for nutrient (particularly nitrogen) recycling, would be detrimental for perennial cropping and would increase fertiliser requirements [69]. In conclusion, the methane potentials used in this study for both sorghum and miscanthus were reliable but remain within the lower range of values published in the same type of study. Consequently, pretreatments prove to be beneficial: indeed the lower the methane potential, the higher the potential impact of pretreatment [70].

Pretreatment results on sorghum revealed higher efficiency values for soda (13% increase of the methane potential) than lime (+ 6%). This was previously observed by Jiang et al. (2017) on giant reeds as well as Nkongndem Nkemka et al. (2016) with *Miscanthus x giganteus* [71, 72]. However, sodium discharge might be environmentally harmful as it can lead to negative impacts such as soil salinization. Lime pretreatment should therefore be preferred [73]. The stronger increase in the miscanthus methane potential (+22% by CaO) in comparison to sorghum (+6% by CaO and 13% by NaOH) could be explained by the higher lignin content in miscanthus. However, these methane potential improvements remain within the same order of magnitude as several previous studies. For example, the *Miscanthus x giganteus* methane potential increased by 20% after a CaO pretreatment [72] and the sorghum potential rose from 8 to 19 % according NaOH pretreatment conditions [74].

Finally, sorghum was shown to outperform miscanthus for mainly two reasons: (1) sorghum has a 58% better BMP compared to miscanthus (2) sorghum contains 70% moisture (versus

15% for miscanthus) and, as digestate is allocated according to the input mass, one megajoule of produced heat is credited of more avoided impact due to digestate spreading.

It is difficult to compare the results of this study to previous work for the main reason that no paper was found about the miscanthus anaerobic digestion with valorization of methane into the gas grid. Yet, the purification and injection of methane represent 40% of the impact on the average. Kiesel et al. (2017) and Wagner et al. (2019) benchmarked the production of electricity by cogeneration in comparison to the German electric mix [19, 62]. They both considered higher miscanthus and biogas yields (18 to 21 t DM ha^{-1}; 229 to 261 Nm^3 CH_4 t^{-1} DM) but no dLUC nor iLUC. Compared to the most similar scenario of this study (HPL_No-LUC_CaO pretreatment), results are in line on the climate change performances of energy production from biomethane based miscanthus. Regarding the other impact categories, despite both study results are not always in agreement (*e.g.*, marine eutrophication is to the advantage of miscanthus in one case and not in the other), they both show that the miscanthus alternative could have an added value compared to the conventional production of energy. These favorable results for miscanthus in Kiesel et al. (2017) and Wagner et al. (2019) are due to several reasons: (1) the overall biogas yield was twice higher than in the present study (2) these studies are not penalized by the injection/purification step (3) the conventional scenario was partially based on much more polluting alternatives (hard coal and oil) than in the present study (natural gas) [19, 62]. On the contrary, the study of Blengini et al. (2011) focused on the cogeneration in the Italian context without taking iLUC into account led to the same overall conclusion as the present study (added value on climate change, but not on the other impact categories)[75]. This study took into account an overall biogas yield only 40% higher. Finally, in a consequential LCA, Tonini et al. (2012) considered dLUC and iLUC in the Danish frame and a 10% higher methane yield [52]. They concluded that the cogeneration of miscanthus-based methane had an added value only on the phosphor-based eutrophication and was worse on the climate change. The comparison for the electricity was, here again, based on hard coal what is probably the most favorable scenario for the miscanthus alternative.

Regarding sorghum, only one study with similar system boundaries up to the injection and combustion of biomethane for heat production has been found [76]. This study considered a similar overall biomethane yield (12 t DM ha^{-1} and 264 Nm^3 CH_4 t^{-1} DM) compared to scenario Main_Int_South without iLUC. As in the present study, without taking into account iLUC, sorghum-based biomethane led to a reduction of the climate change impact and, depending on the categories, to an increase or a decrease of the impacts. Buratti et al. (2013) also studied the injection of sorghum biomethane in the grid to be used as vehicle fuel in comparison to gasoline, diesel or natural gas, but unfortunately focused the assessment on the climate change impact [77]. As in the present work, they concluded that sorghum-based methane, without taking into iLUC, is better than natural gas. Blengini et al. (2011) compared the sorghum biogas cogeneration to the natural gas-based electricity and heat production [75]. Compared to the most comparable scenario of this study (Main_Int_South without iLUC), the overall methane production was 2.2 times higher (23 t DM ha^{-1} and 297 Nm^3 CH_4 t^{-1} DM) but conclusions were similar to the ones presented here (better on climate change but worse on photochemical ozone formation, acidification and marine eutrophication). Agostini et al. (2015) confirmed that taking into account iLUC may overcome the benefit of using sorghum-based methane to produce heat and electricity [78]. Indeed, despite a 1.4 higher production of methane (17 t DM ha^{-1} and 256 Nm^3 CH_4 t^{-1} DM), the climate change impact became equivalent or slightly higher than the conventional alternative. Regarding other impact categories, the sorghum scenario had an added value only on the fossil-resource depletion. Finally, for both sorghum and miscanthus, methane yield was assumed to be 85% of BMP values. In contrast, published studies generally do not mention such a factor considering lower methane production in a full-scale digester and may overestimate methane yields by considering BMP values.

Conclusion

For both sorghum and miscanthus, the contribution analysis reveals that both the yield and agricultural practices have the highest influence. Next, the purification and injection step contributes strongly because of the infrastructure, electricity consumption and methane losses. The digestate spreading and use as fertiliser also represents a key source of pollution through nitrogen emissions. Lime pretreatment entails a lower methane production enhancement than soda but is more favourable because its production generates significantly less impacts. For miscanthus, this doesn't modify the conclusion of the impact analysis but improves the single score of the scenario where miscanthus is cultivated on a marginal and high productive land in comparison with natural gas. In the case of sorghum, the conclusion of the impact analysis is not modified but the single score of all sorghum scenarios (exepted iLUC) are equivalent or higher than for natural gas. In the studied scenarios, the high iLUC impact was not or hardly compensated by environmental benefits of biomass-based methane. This highlights the importance of using catch crops, or long lifespan periennal crops (at least 20 years), or to ensure that land is still cultivated for energy production after 20 years of exploitation, or in the case of dedicaced annual crops, the necessity of achieving high biomas and biomethane yields. However, single score results have been found to be in favour of sorghum-based methane, which can thus be an option to mitigate climate change. In addition, the growth of biomass for anaerobic digestion or other applications needs to be monitored to avoid potential risks such as iLUC or loss in biodiversity.

Acknowledgements

Acknowledgements are addressed to David Pot and Stéphanie Arnould and Maryse Brancourt-Humel who provided sorghum and miscanthus samples, respectively. Authors are also grateful to the agriculture specialist panels for their help in building the scenarios: Alain Besnard and Sylvain Marsac from Arvalis, Pierre Malvoisin from Aelred, Thierry Jacquet, Magali Berthou and Claire Brami from Phytorestore, Sylvain Frédéric from Naskeo-Environnement, David Pot from CIRAD, Chantal Loyce from AgroParisTech and Dominique Romelot from Axereal.

Declarations

Funding

The authors acknowledge the French National Research Agency for funding the "Biomass For the Future" project (ANR, grant ANR-11-BTBR-0006-BFF).

Conflicts of interest/Competing interests

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

Availability of data and material (data transparency)

Data will be available on request

Code availability

Not applicable

Author's contributions

CJ Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – Original Draft, Writing – Review & Editing

HLT Methodology, Validation, Formal analysis,
Investigation, Writing – Original Draft, Writing – Review & Editing

HC Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – Original Draft, Writing – Review & Editing- Funding acquisition

Abbreviations

Acidif.	Acidification
AD	Anaerobic digestion
BFF	Biomass for the Future project
CHP	Combined heat and power
Clim. Chang. Excl.	Climate change excluding biogenic carbon
dLUC	Direct consequences of Land Use Change
DM	Dry matter
Ecotox. Freshwat.	Freshwater ecotoxicity
Eutr. Freshwat.	Freshwater eutrophication
Eutr. Marine	Marine eutrophication
Eutr. Terres.	Terrestrial eutrophication
HHV	Higher heating value
GHG	Green house gas
HPL	Higher productive land
Hum. tox. cancer	Human toxicity cancer
Hum. tox. non-cancer	Human toxicity non-cancer

ILCD	International Life Cycle Database
iLUC	Indirect consequences of Land Use Change
Ion. rad.	Ioniozing radiation
OM	Organic matter
Oz. Depl.	Ozone depletion
Part. Mat. Form.	Particulate matter formation
Photoch. ozone form.	Photochemical ozone formation
SOC	Soil Organic Carbon
SS	Solid state
Water dep.	Water depletion
Min., foss. & renew. Depl.	Mineral, fossil and renewable depletion

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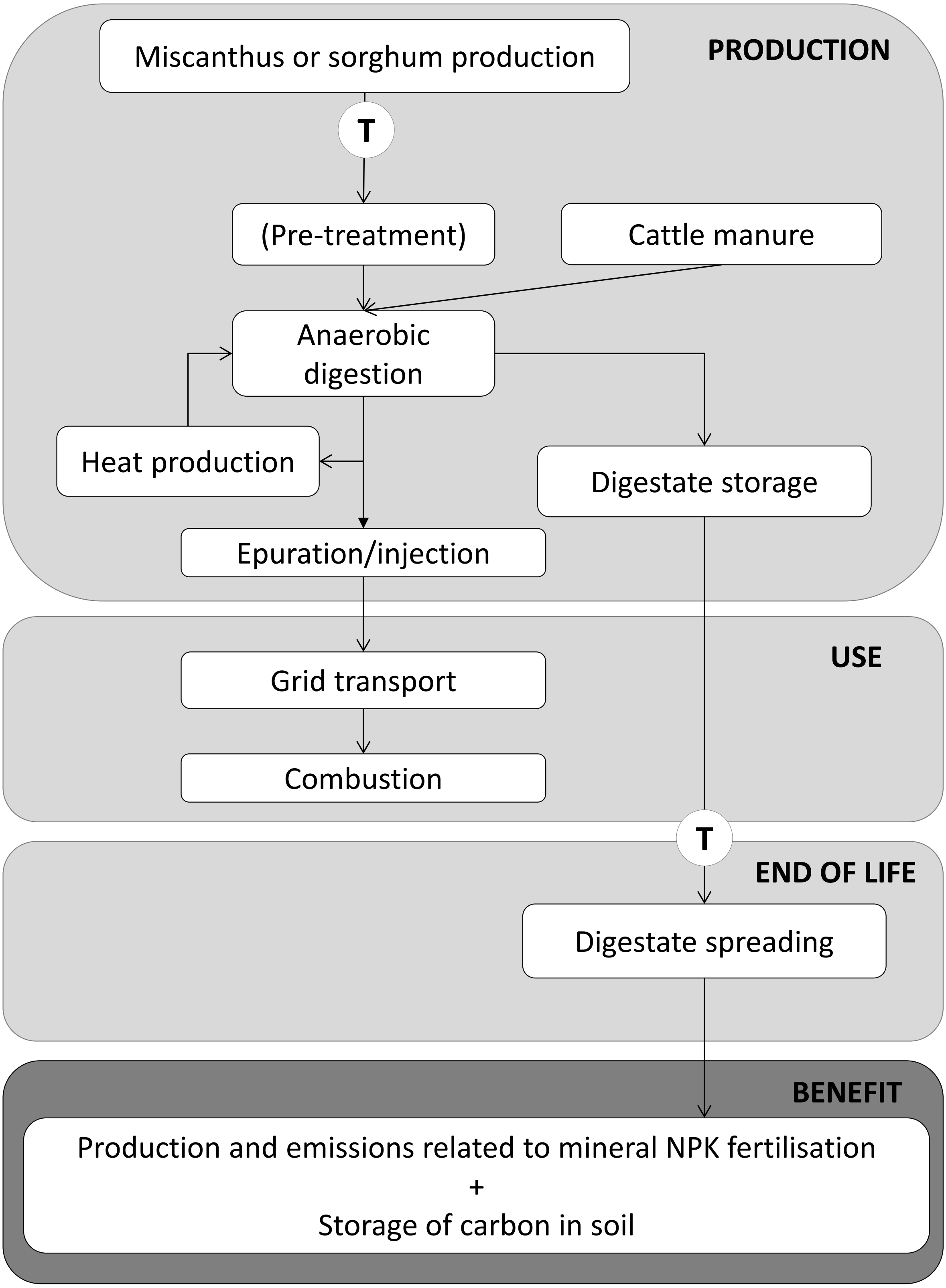
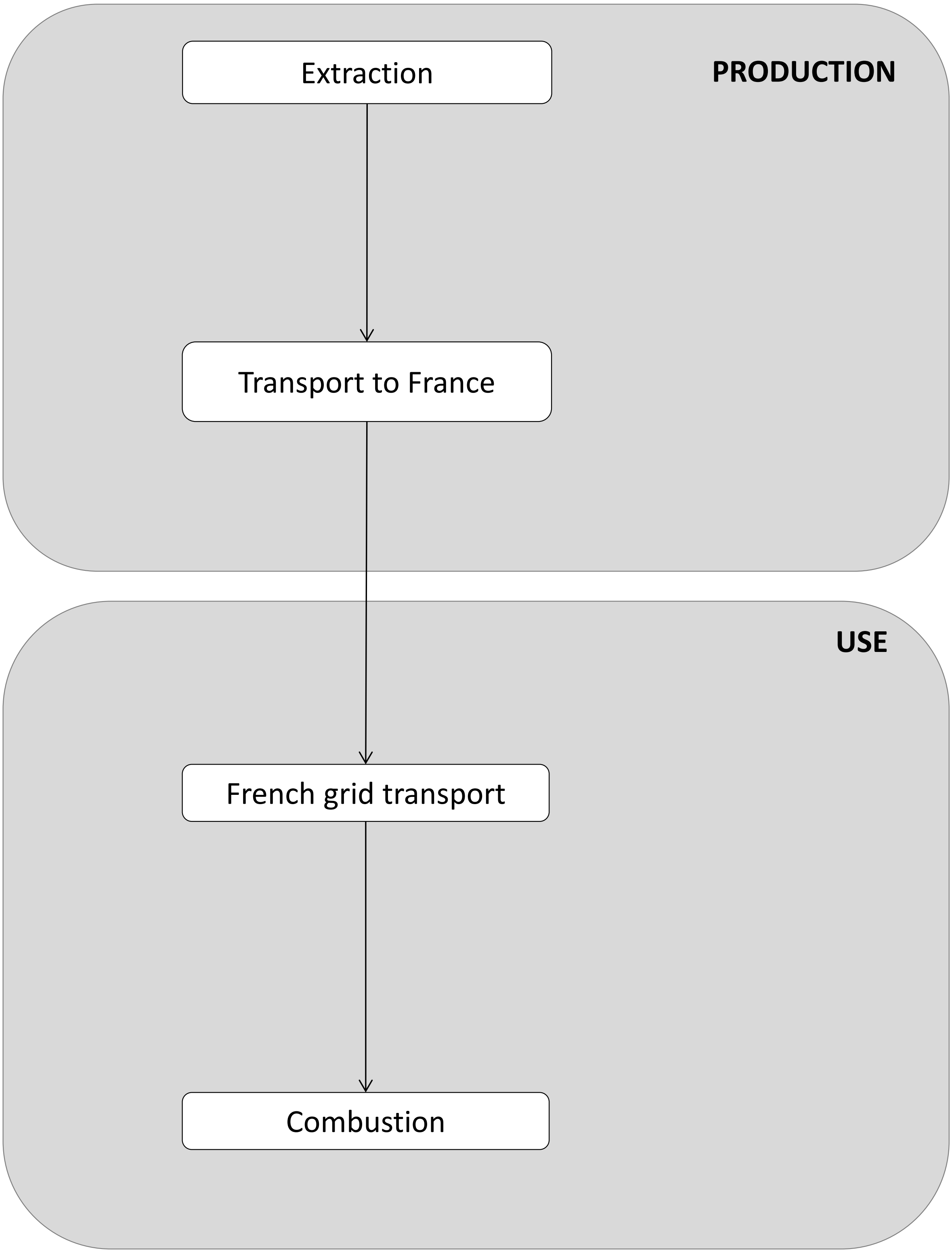
Fig.1: System boundaries for (a) the sorghum or miscanthus based scenario (b) natural gas



(a)

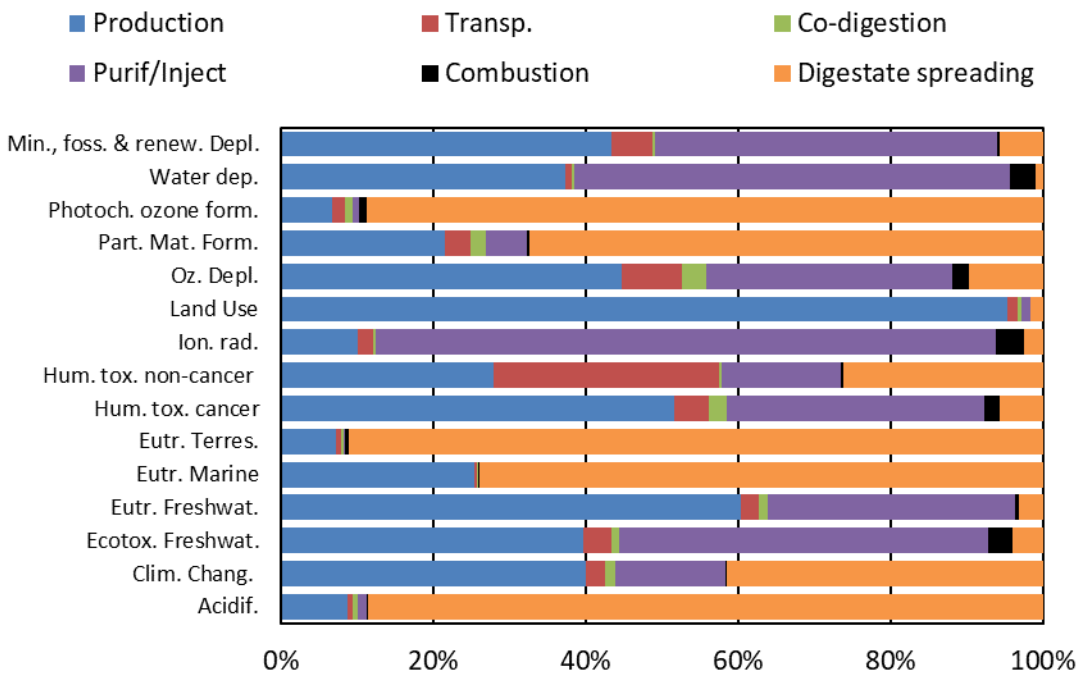


Extraction
PRODUCTION
Transport to France
USE
French grid transport
Combustion

(b)

Fig. 2: Contribution analysis of the heat production from miscanthus based methane (a) without pretreatment (b) with CaO pretreatment (c) with NaOH pretreatment

(a)



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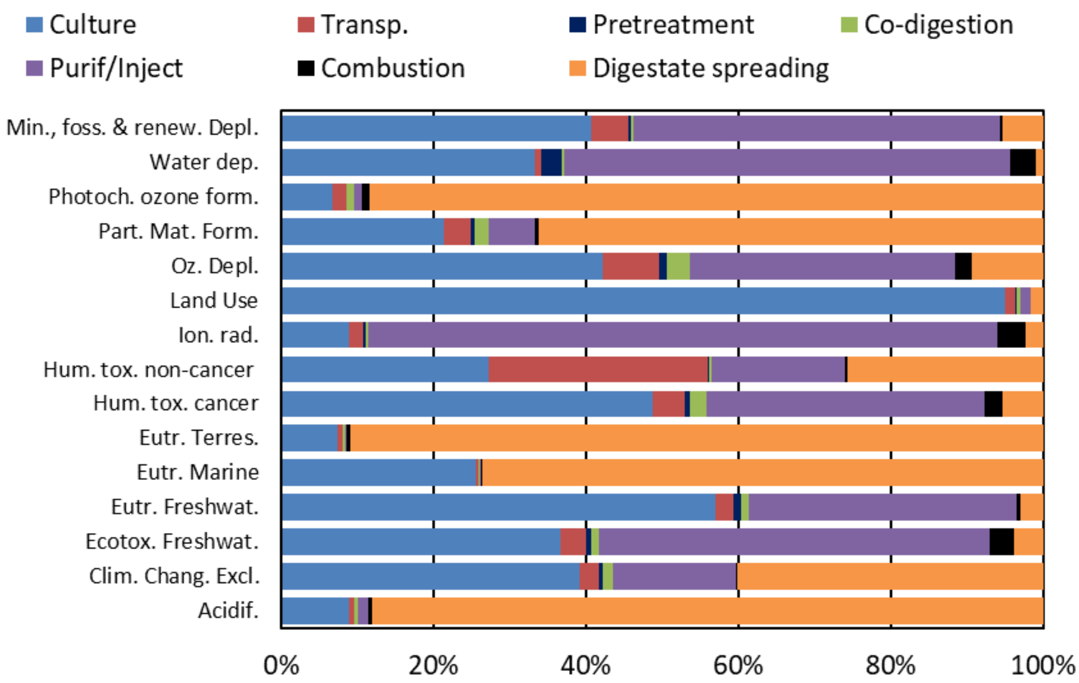
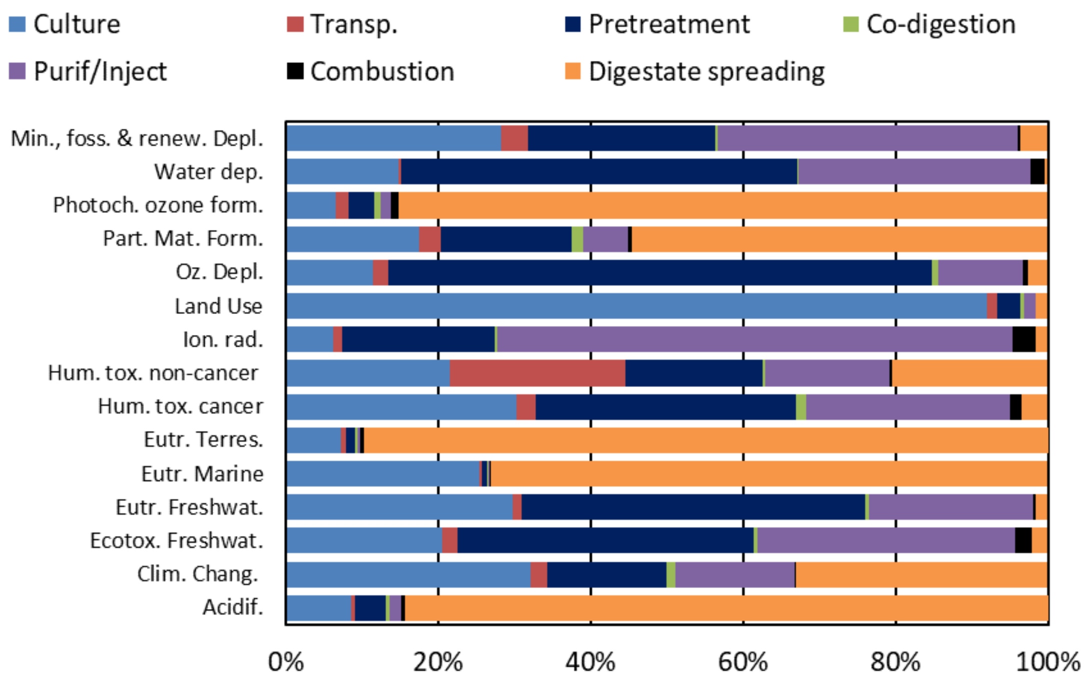


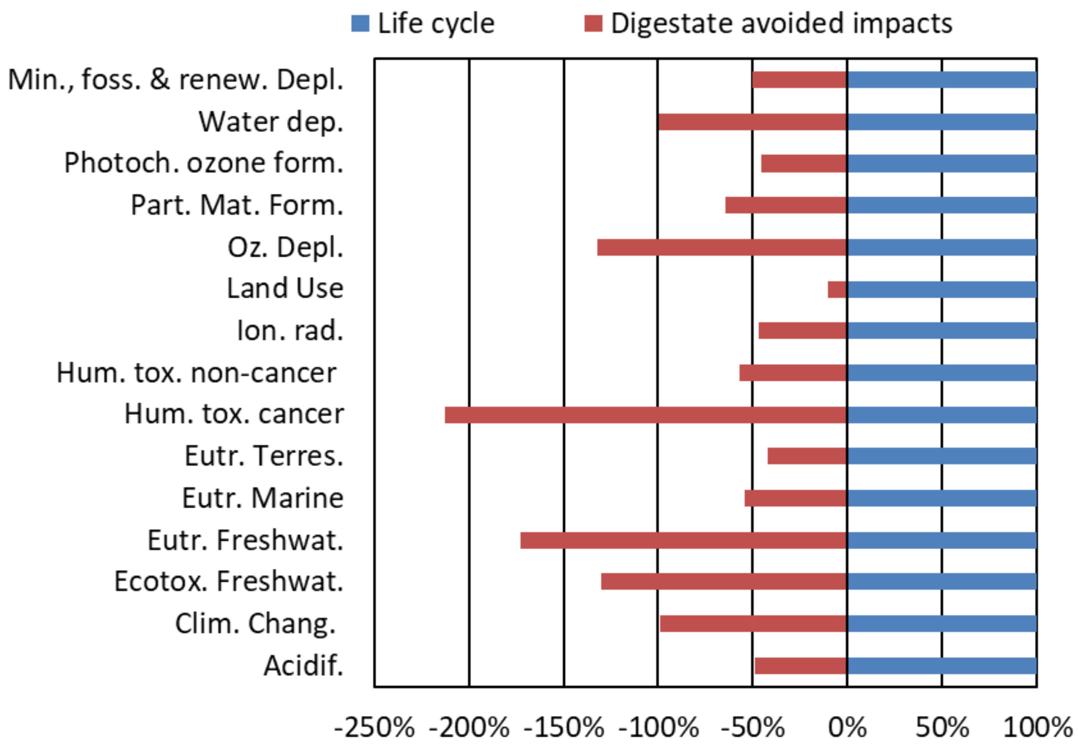
(c)



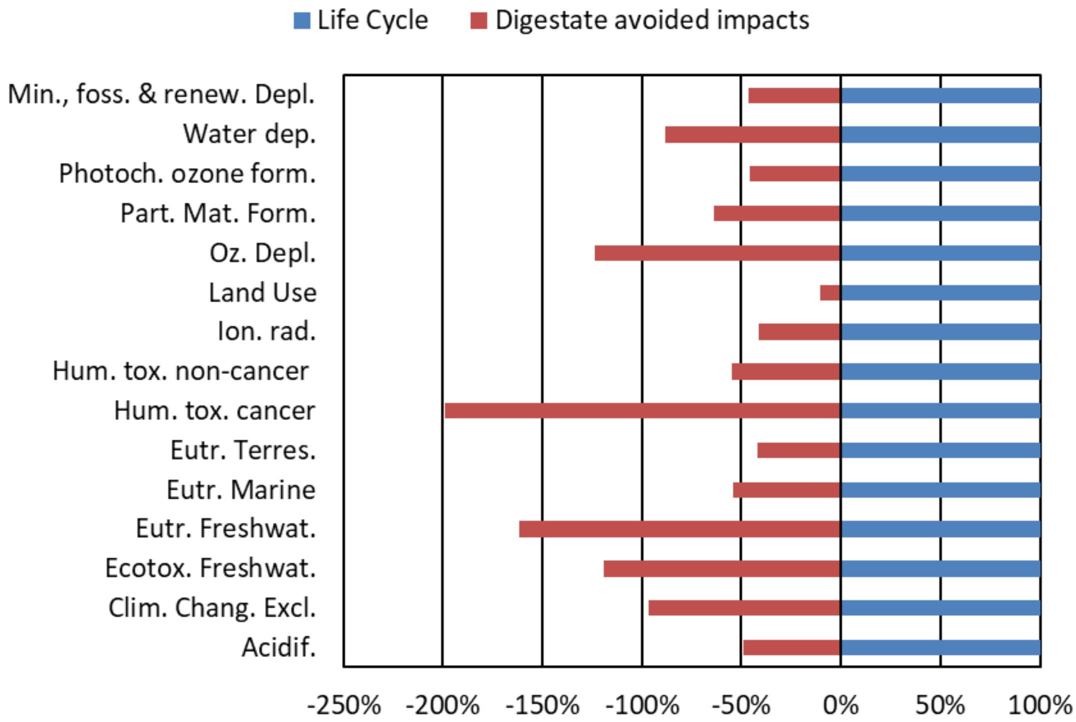
Culture
Transp.
Pretreatment
Co-digestion
Purif/Inject
Combustion
Digestate spreading
Min., foss. & renew. Depl.
Water dep.
Photoch. ozone form.
Part. Mat. Form.
Oz. Depl.
Land Use
Ion. rad.
Hum. tox. non-cancer
Hum. tox. cancer
Eutr. Terres.
Eutr. Marine
Eutr. Freshwat.
Ecotox. Freshwat.
Clim. Chang.
Acidif.
0%
20%
40%
60%
80%
100%

Fig.3: Influence of the avoided impact due to sorghum co-digestion digestate spreading (a) without pretreatment (b) with CaO pretreatment (c) with NaOH pretreatment

(a)



(b)


Life Cycle
Digestate avoided impacts
Min., foss. & renew. Depl.
Water dep.
Photoch. ozone form.
Part. Mat. Form.
Oz. Depl.
Land Use
Ion. rad.
Hum. tox. non-cancer
Hum. tox. cancer
Eutr. Terres.
Eutr. Marine
Eutr. Freshwat.
Ecotox. Freshwat.
Clim. Chang. Excl.
Acidif.
-250% -200% -150% -100% -50% 0% 50% 100%

(c)

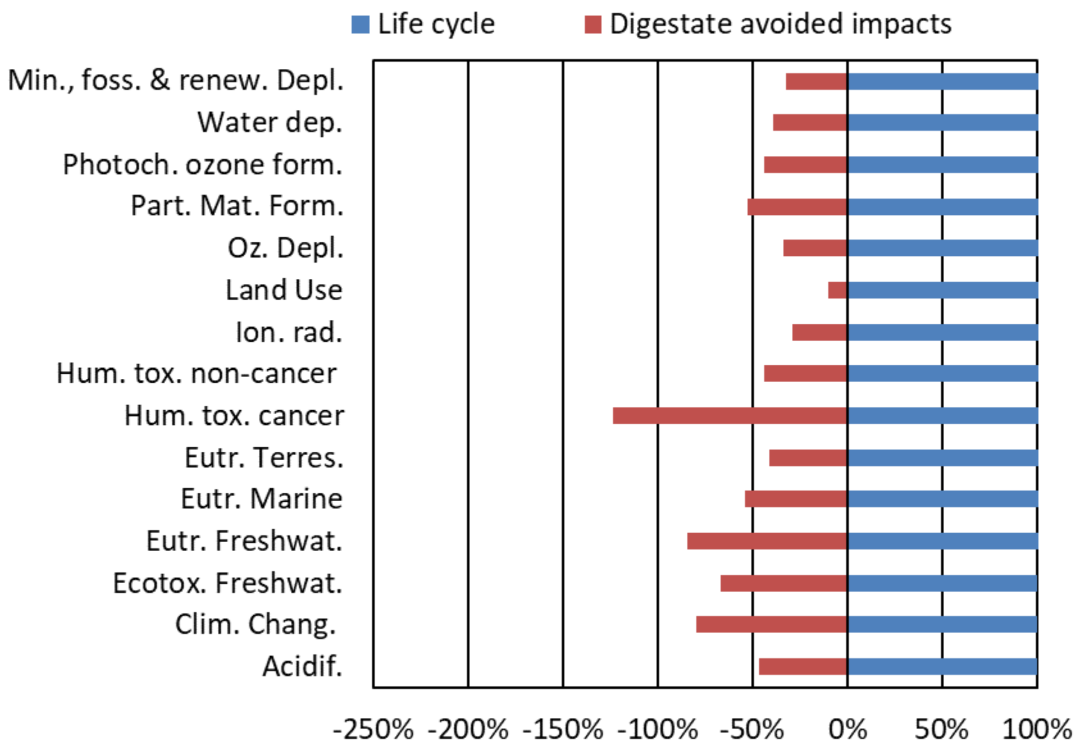
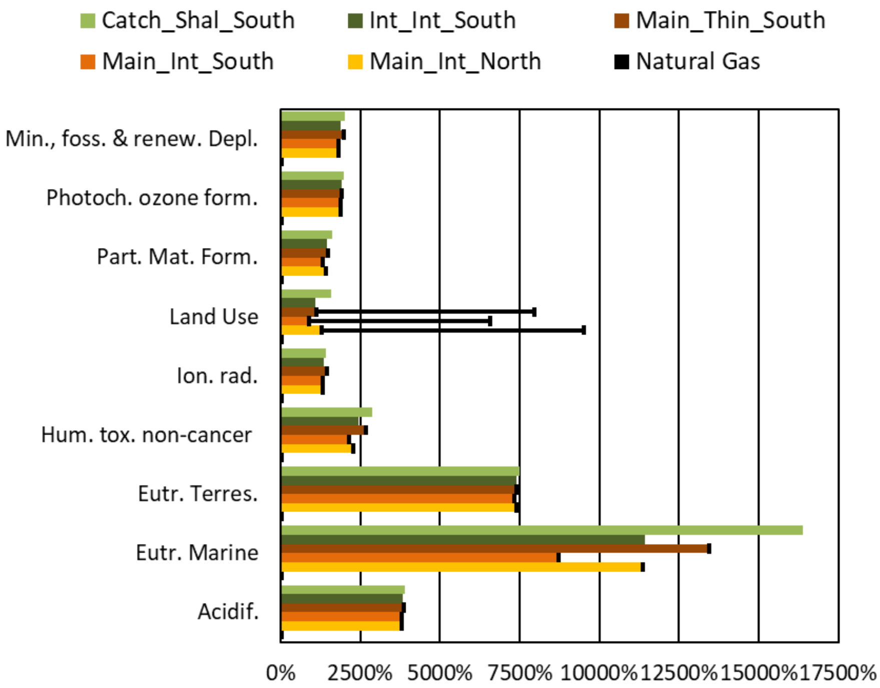

Life cycle
Digestate avoided impacts
Min., foss. & renew. Depl.
Water dep.
Photoch. ozone form.
Part. Mat. Form.
Oz. Depl.
Land Use
Ion. rad.
Hum. tox. non-cancer
Hum. tox. cancer
Eutr. Terres.
Eutr. Marine
Eutr. Freshwat.
Ecotox. Freshwat.
Clim. Chang.
Acidif.
-250% -200% -150% -100% -50% 0% 50% 100%

Fig. 4: Comparison of heat produced from sorghum-based methane without pretreatment to heat produced from natural gas. Error bars show impacts when iLUC is considered. (a) impacts in favour of natural gas and (b) impacts in favour of sorghum-based methane.

(a)



(b)

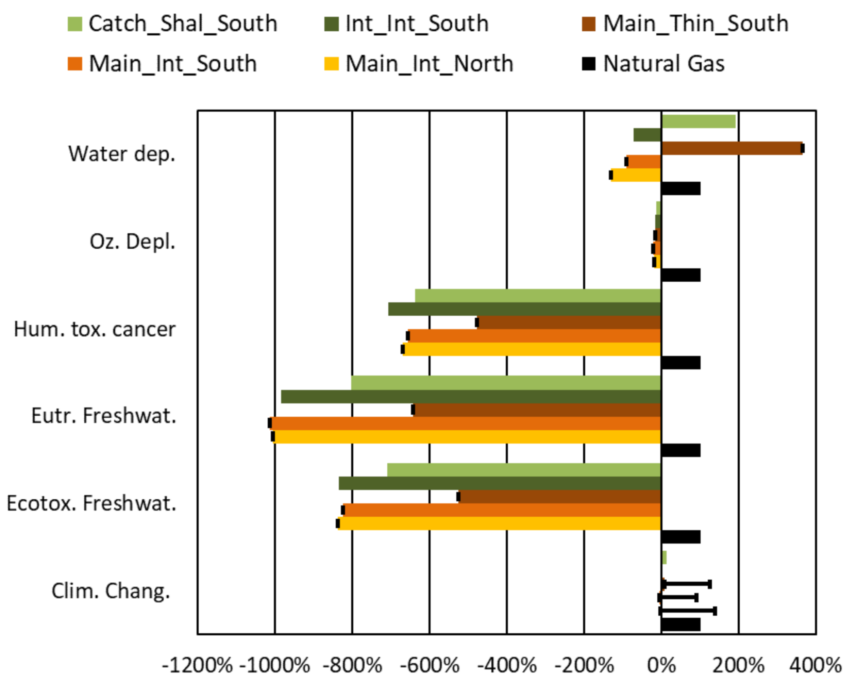
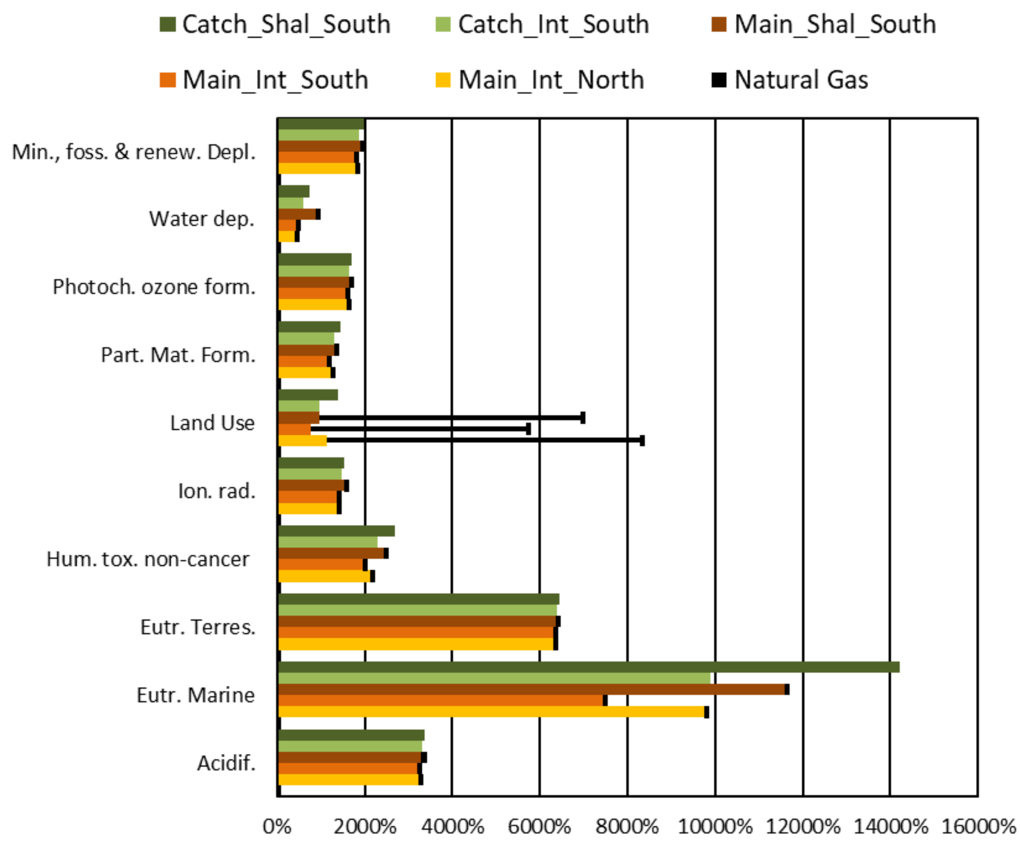


Fig. 5: Comparison of heat produced from sorghum-based methane with lime pretreatment to heat produced from natural gas. Error bars indicate impacts when iLUC is considered. (a) impacts in favour of natural gas and (b) impacts in favour of sorghum-based methane.

(a)



(b)

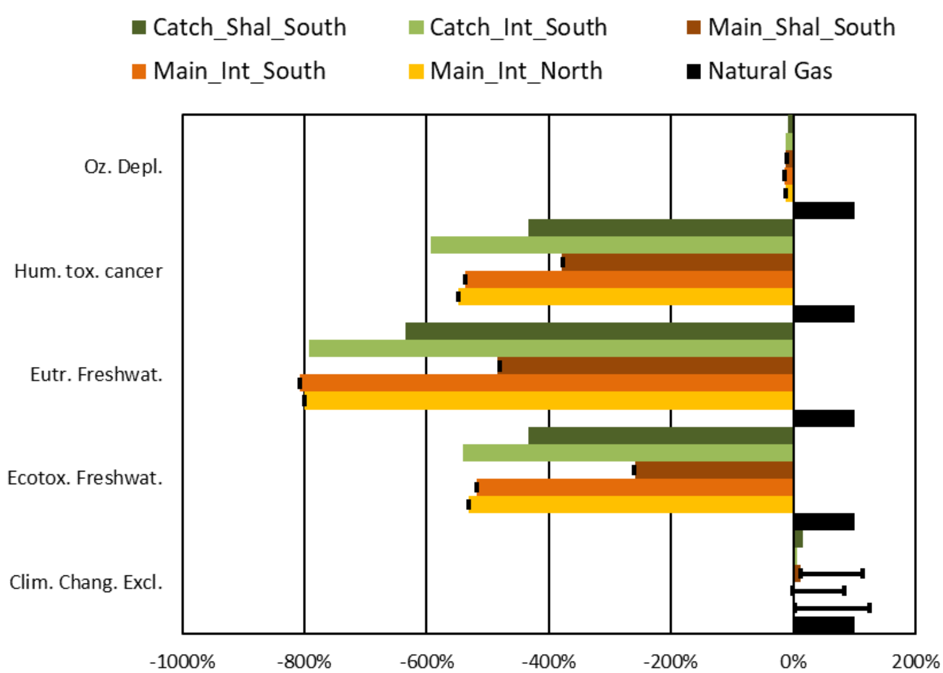
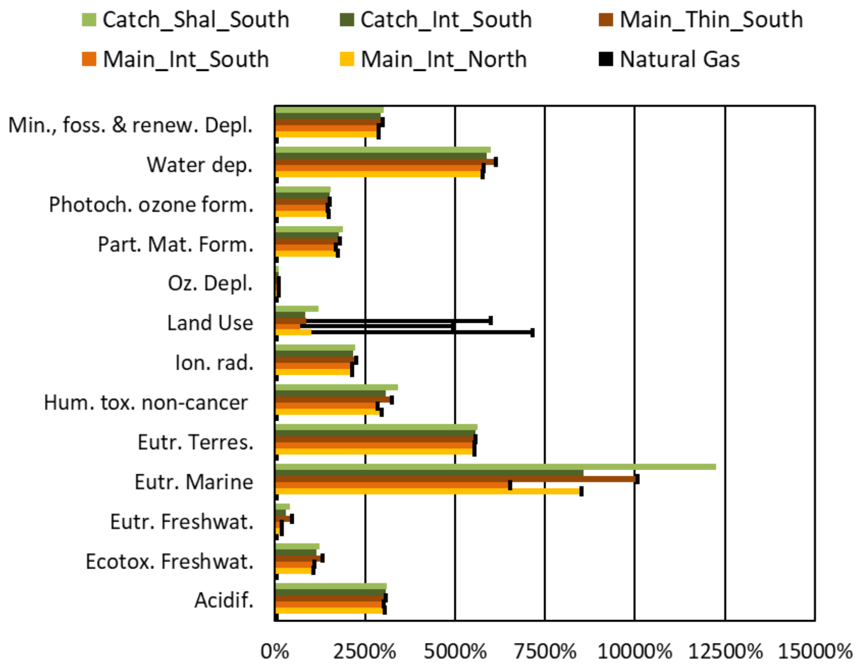


Fig. 6: Comparison of heat produced from sorghum-based methane with a sodium hydroxide pretreatment to heat produced from natural gas. Error bars show impacts when iLUC is considered. (a) impacts in favour of natural gas and (b) impacts in favour of sorghum-based methane.

(a)



(b)

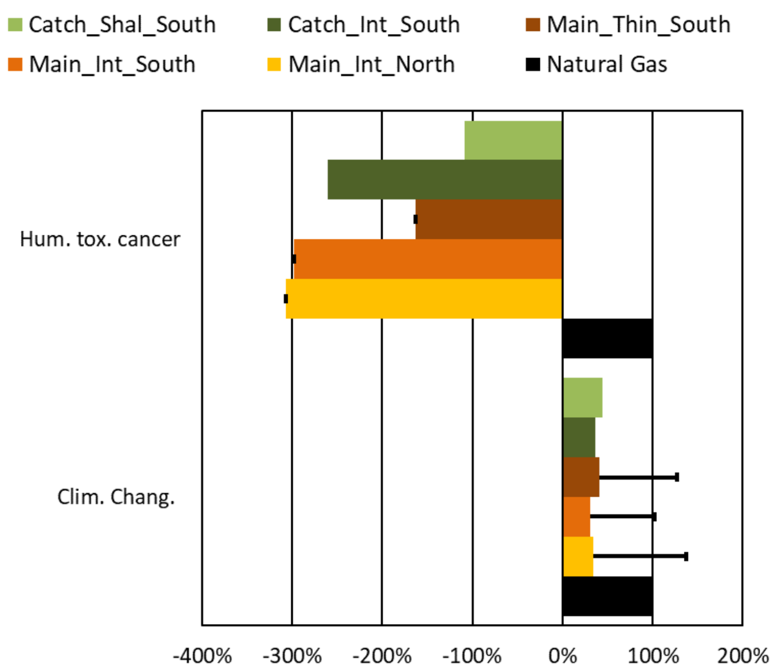
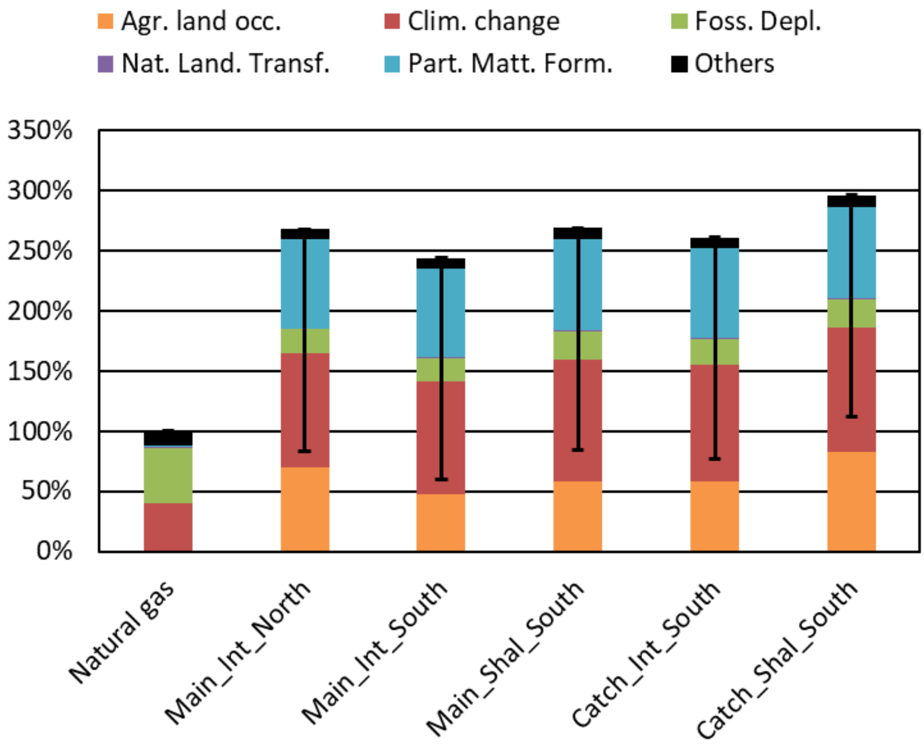
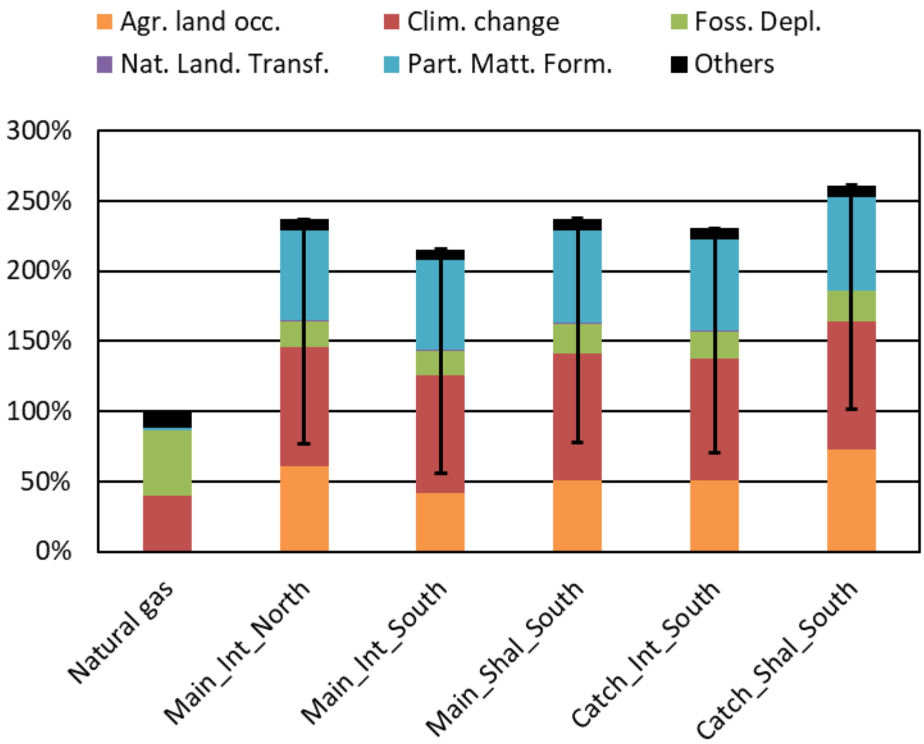


Fig. 7: Comparison of heat produced from sorghum based methane to heat produced from natural gas (ReCiPe HA). No iLUC considered. Error bars represent avoided impacts due to digestate use for fertilisation. The upper part of the histogram shows the final single score without avoided impacts due to digestate use and the lower part of the error bar the final single score when the avoided impacts due to digestate use are taken into account. (a) without pretreatment. (b) CaO pretreatment (c) NaOH

(a)

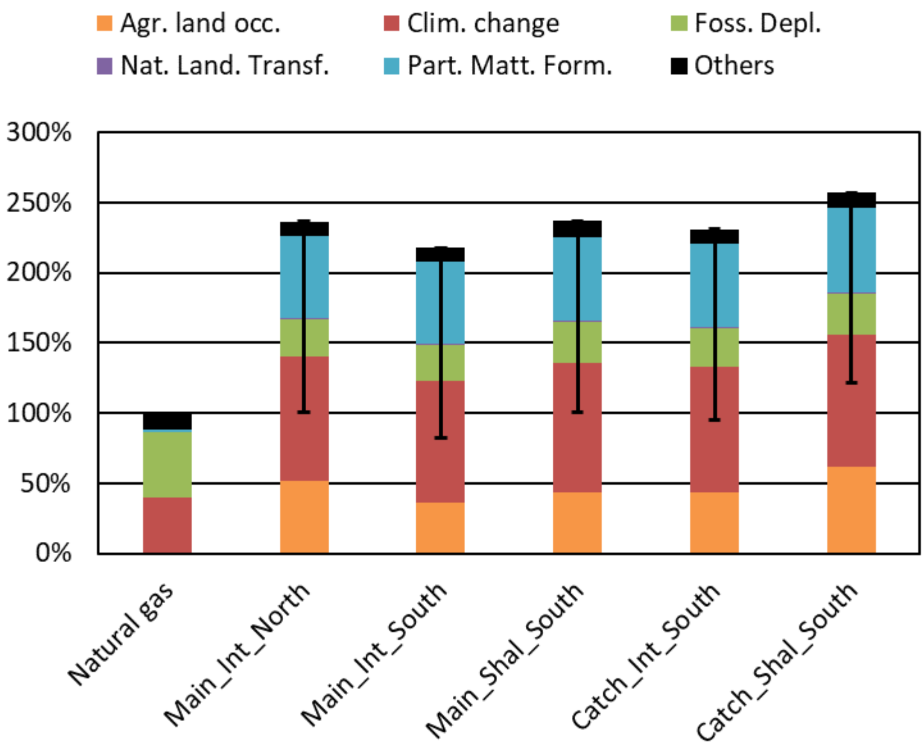


(b)



Agr. land occ.
Clim. change
Foss. Depl.
Nat. Land. Transf.
Part. Matt. Form.
Others
300%
250%
200%
150%
100%
50%
0%
Natural gas
Main_Int_North
Main_Int_South
Main_Shal_South
Catch_Int_South
Catch_Shal_South

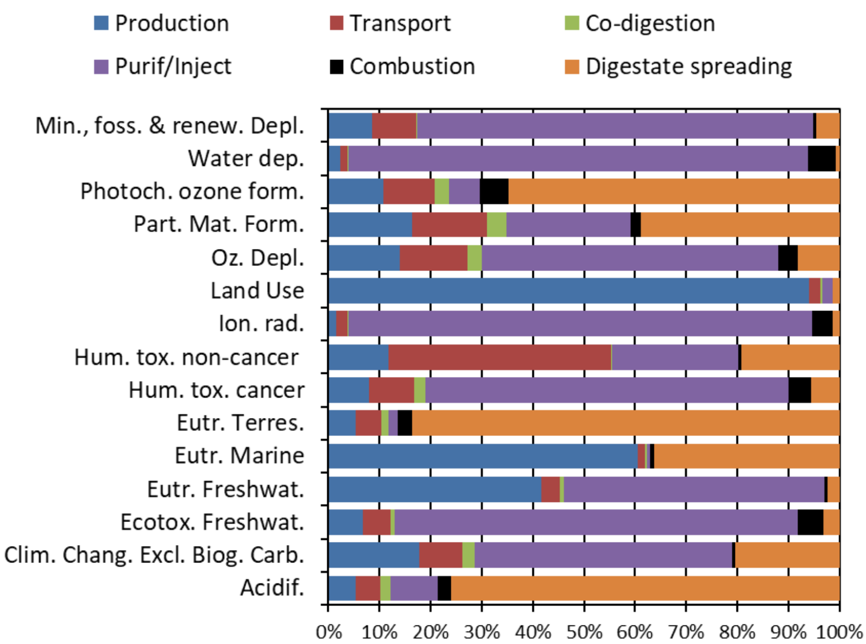
(c)



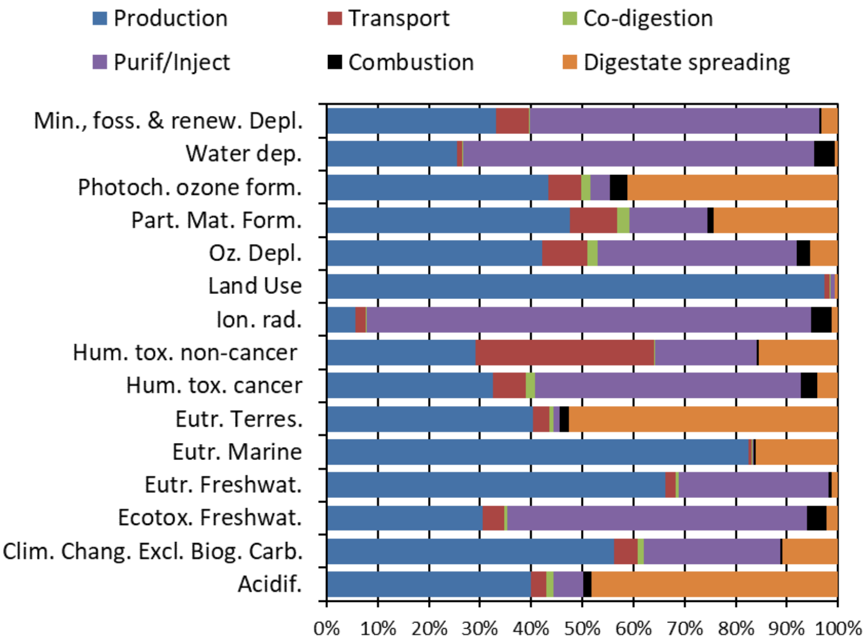
Agr. land occ.
Clim. change
Foss. Depl.
Nat. Land. Transf.
Part. Matt. Form.
Others
300%
250%
200%
150%
100%
50%
0%
Natural gas
Main_Int_North
Main_Int_South
Main_Shal_South
Catch_Int_South
Catch_Shal_South

Fig. 8: Contribution analysis for the miscanthus (a) HPL no-pretreatment (b) LPL no-pretreatment (c) HPL lime pretreatment (d) LPL lime pretreatment

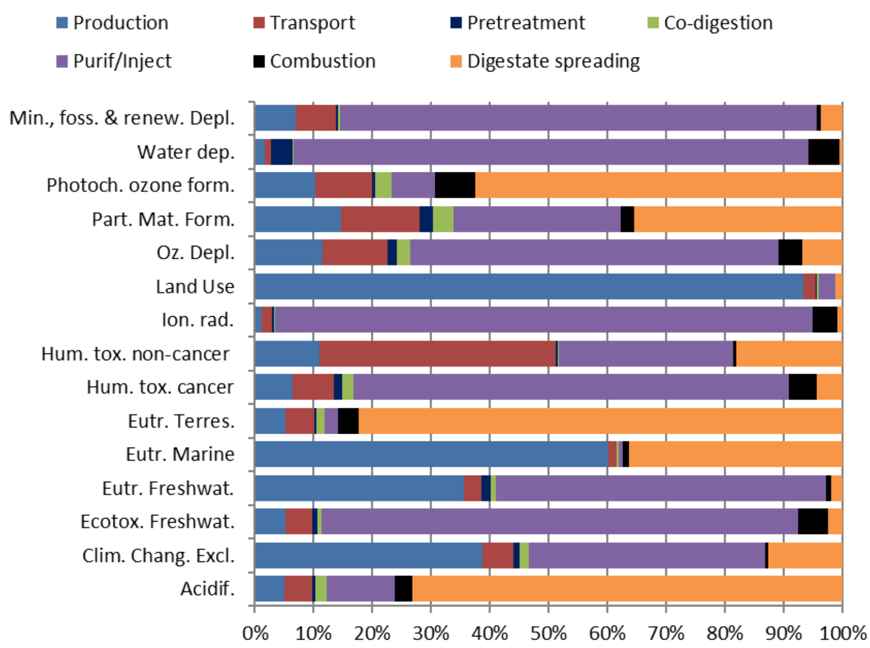
(a)



(b)

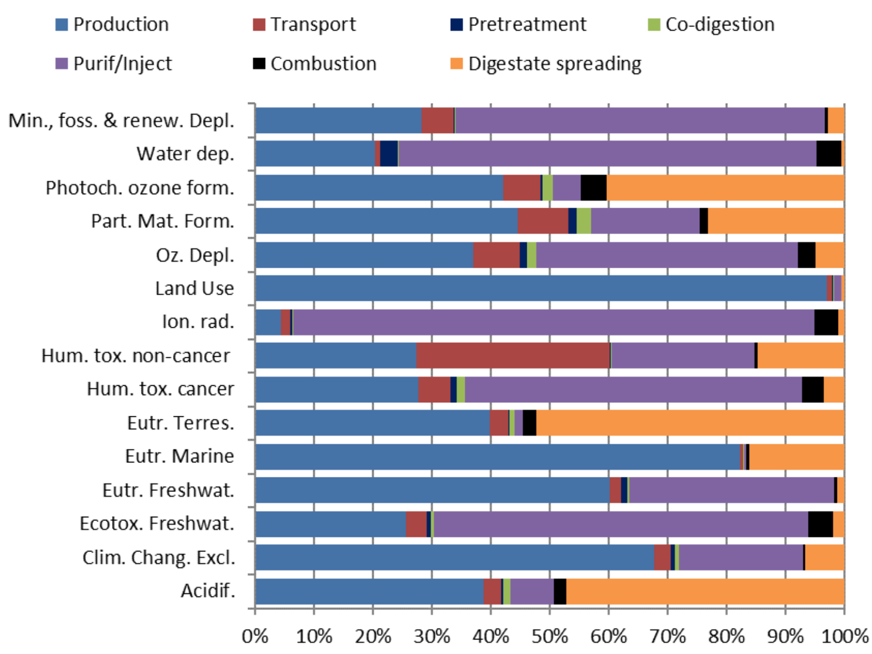


(c)



Production
Transport
Pretreatment
Co-digestion
Purif/Inject
Combustion
Digestate spreading
Min., foss. & renew. Depl.
Water dep.
Photoch. ozone form.
Part. Mat. Form.
Oz. Depl.
Land Use
Ion. rad.
Hum. tox. non-cancer
Hum. tox. cancer
Eutr. Terres.
Eutr. Marine
Eutr. Freshwat.
Ecotox. Freshwat.
Clim. Chang. Excl.
Acidif.
0%
10%
20%
30%
40%
50%
60%
70%
80%
90%
100%

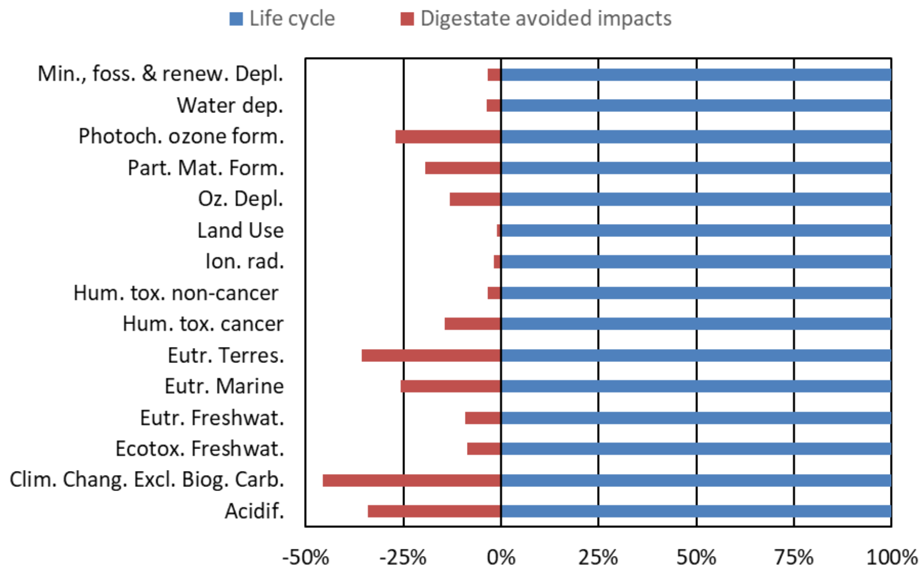
(d)



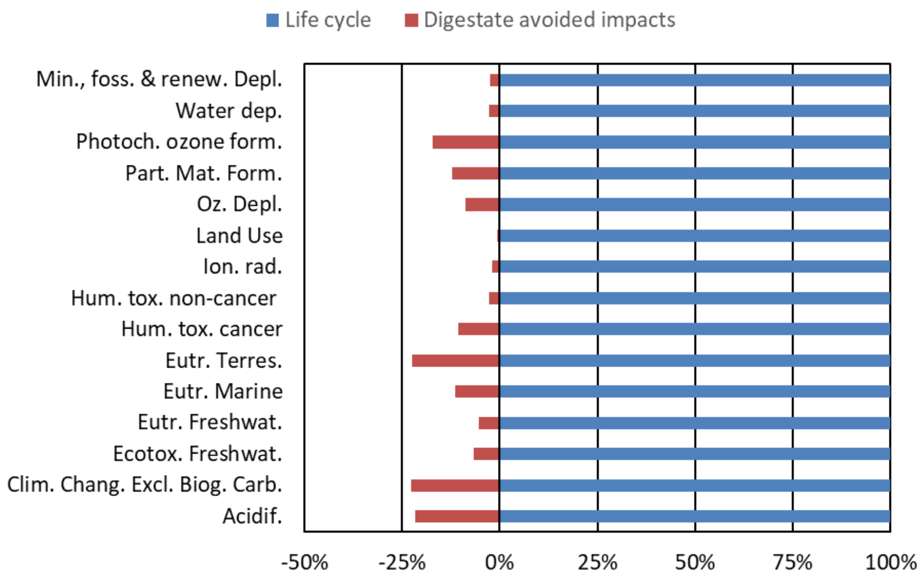
Production
Transport
Pretreatment
Co-digestion
Purif/Inject
Combustion
Digestate spreading
Min., foss. & renew. Depl.
Water dep.
Photoch. ozone form.
Part. Mat. Form.
Oz. Depl.
Land Use
Ion. rad.
Hum. tox. non-cancer
Hum. tox. cancer
Eutr. Terres.
Eutr. Marine
Eutr. Freshwat.
Ecotox. Freshwat.
Clim. Chang. Excl.
Acidif.
0%
10%
20%
30%
40%
50%
60%
70%
80%
90%
100%

Fig. 9: Influence of the digestate spreading (a) HPL no-pretreatment (b) LPL no-pretreatment (c) HPL lime pretreatment (d) LPL lime pretreatment

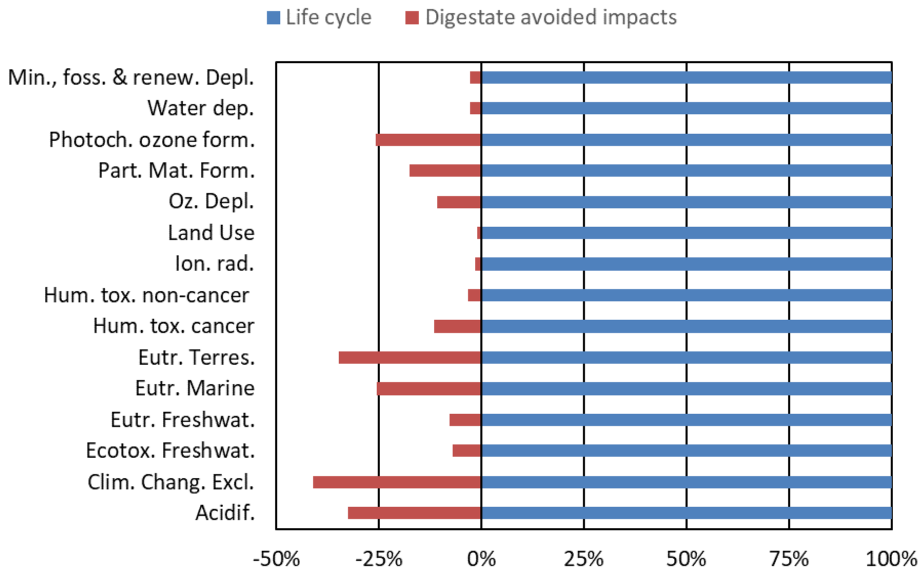
(a)



(b)



(c)


Life cycle
Digestate avoided impacts
Min., foss. & renew. Depl.
Water dep.
Photoch. ozone form.
Part. Mat. Form.
Oz. Depl.
Land Use
Ion. rad.
Hum. tox. non-cancer
Hum. tox. cancer
Eutr. Terres.
Eutr. Marine
Eutr. Freshwat.
Ecotox. Freshwat.
Clim. Chang. Excl.
Acidif.
-50%
-25%
0%
25%
50%
75%
100%

(d)

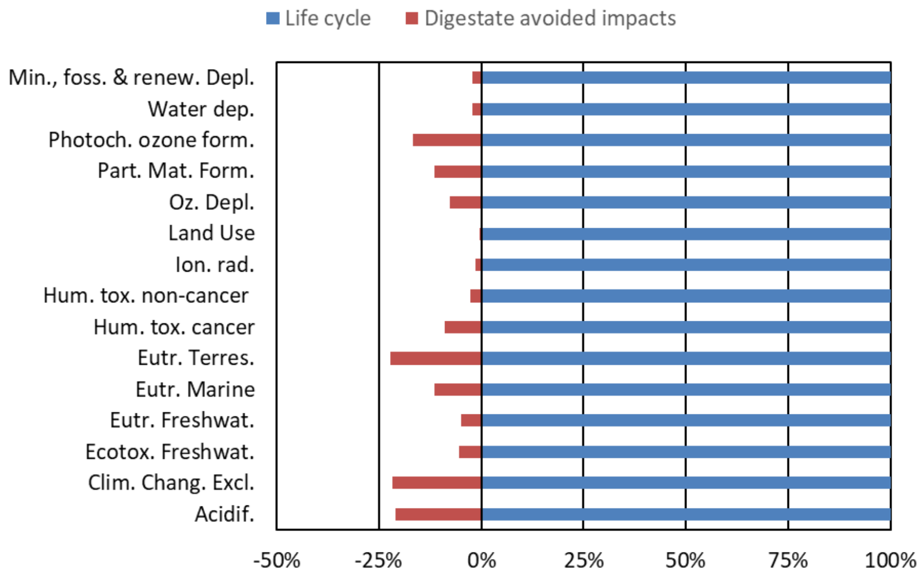
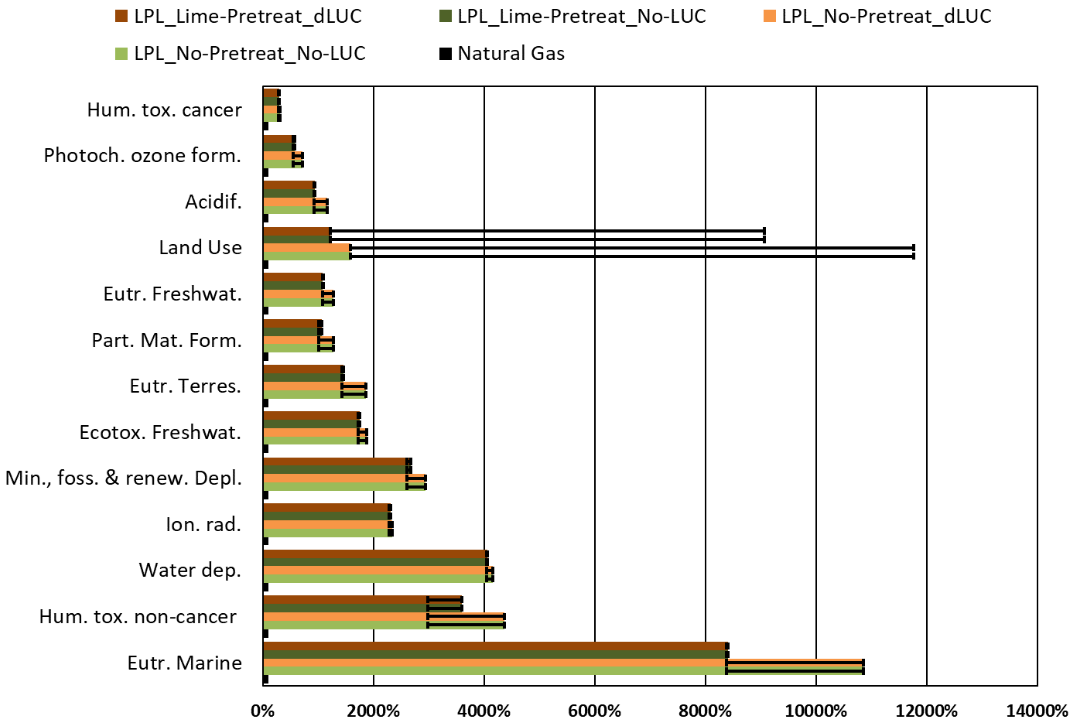

Life cycle
Digestate avoided impacts
Min., foss. & renew. Depl.
Water dep.
Photoch. ozone form.
Part. Mat. Form.
Oz. Depl.
Land Use
Ion. rad.
Hum. tox. non-cancer
Hum. tox. cancer
Eutr. Terres.
Eutr. Marine
Eutr. Freshwat.
Ecotox. Freshwat.
Clim. Chang. Excl.
Acidif.
-50%
-25%
0%
25%
50%
75%
100%

Fig. 10: Comparison of heat produced from miscanthus (LPL) based-methane to heat produced from natural gas. Uncertainty bars represent the influence of cultivating miscanthus on a land in competition with food production: (a) impacts in favour of natural gas and (b) impacts in favour of miscanthus-based methane

(a)



(b)

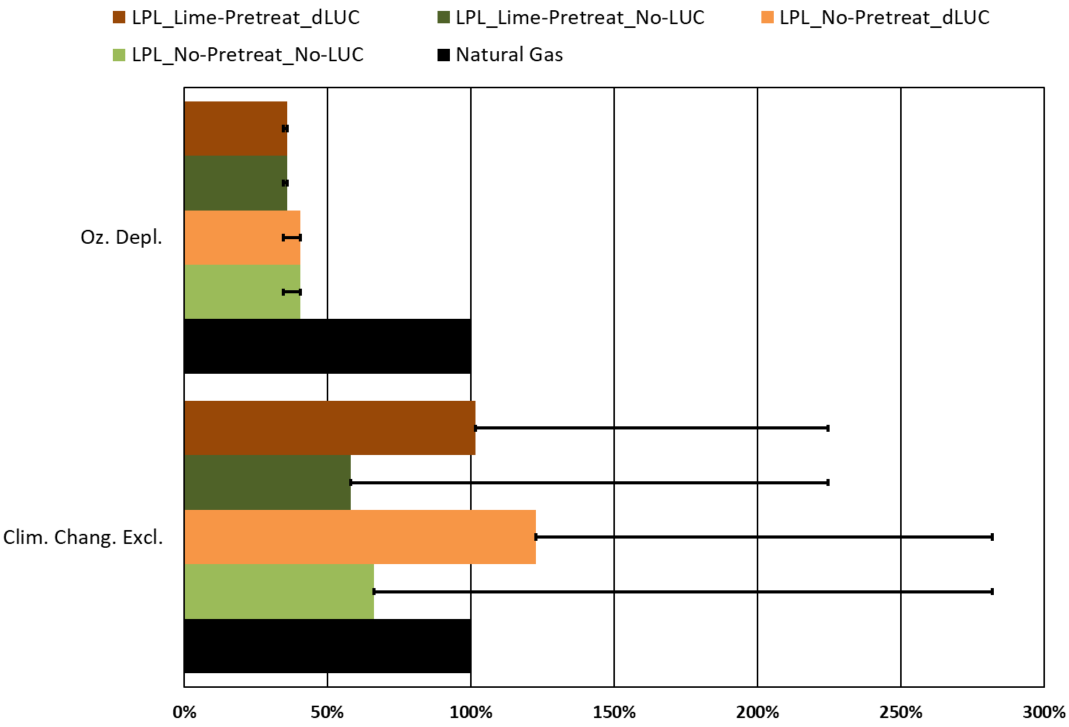
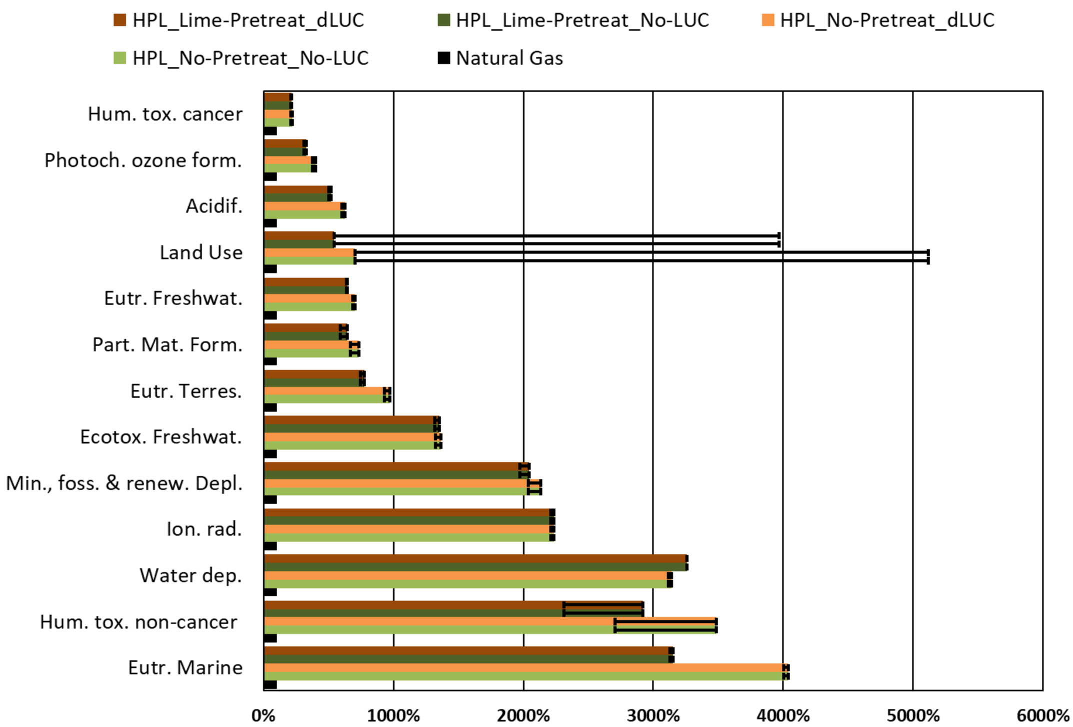

LPL_Lime-Pretreat_dLUC
LPL_Lime-Pretreat_No-LUC
LPL_No-Pretreat_dLUC
LPL_No-Pretreat_No-LUC
Natural Gas
Oz. Depl.
Clim. Chang. Excl.
0%
50%
100%
150%
200%
250%
300%

Fig. 11: Comparison of heat produced from miscanthus (HPL) based-methane to heat produced from natural gas. Uncertainty bars represent the influence of cultivating miscanthus on a land in competition with food production: (a) impacts in favour of natural gas and (b) impacts in favour of miscanthus-based methane

(a)



(b)

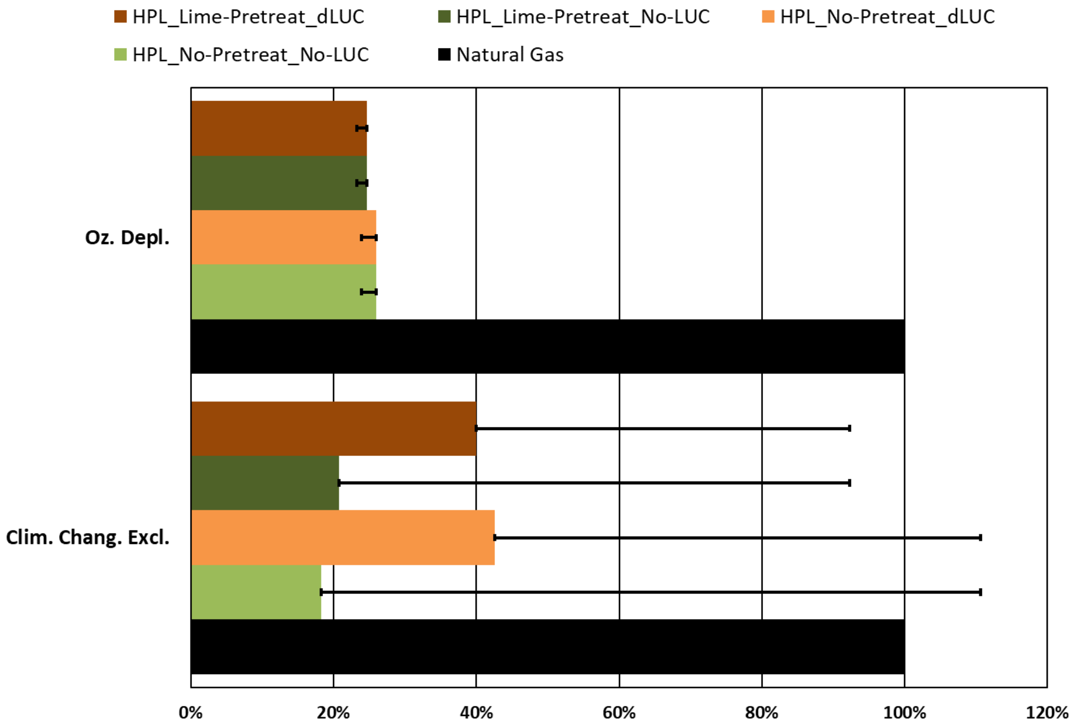


Fig. 12: Comparison of heat produced from miscanthus-based methane to heat produced from natural gas (ReCiPe HA). No iLUC considered. Error bars represent avoided impacts due to digestate use for fertilisation. The upper part of the histogram shows the final single score without avoided impacts due to digestate use and the lower part of the error bar the final single score when the avoided impacts due to digestate use are taken into account.