NoAW project

Innovative approaches to turn agricultural waste into ecological and economic assets

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1. Document Info

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1.2. Revision history

Version	Date	Modified by	Comments
1	November 12, 2020	Stig I. Olsen	Starting version
2	November 24, 2020	Stig I. Olsen	Version 2, after review

1.3. Dissemination level

This deliverable is part of a project that has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 688338	
Dissemination Level	
PU Public	**PU**
CI Classified, as referred to Commission Decision 2001/844/EC	
CO Confidential, only for members of the consortium (including the Commission Services)	

Note that publication should as far as possible be delayed until Nilsson et al, 202x has been published.

2. Summary

Background	In WP2 methodologies have been developed to assess the lab and pilot scale technologies/pathways explored and developed in other WPs of the NoAW project. In WP6 the most promising technologies/pathways are chosen for upscaling experiments and task 6.1 apply methods and results from WP2 to evaluate the environmental implications of the upscaled technologies.
Objectives	To identify the origin and the magnitude of the potential environmental benefits (measured as 18 environmental impact categories, a.o. climate change) of upscaling of the systems investigated in Task 6.2. These are: • PHA production • Solvent based extraction of polyphenols out of wine pomaces • Production of active packaging materials using polyphenols received after solvent based extraction • Utilization of vine shoots (or extraction residues) for production of biocomposites with PHA
Methods	The evaluation has been performed using the TM-LCA (Territorial Metabolism Life Cycle Assessment) and MCDA (Multicriteria decision analysis) tools developed in WP2. It has been applied for upscaled technologies with different types of feedstock and in different regions.
Results & implications	Overall, it can be concluded that biorefinery technologies are environmentally beneficial compared to traditional ways of handling agricultural wastes. Biogas production, especially with AD booster pretreatment, is the environmentally best option in most cases but co-production of PHA also leads to environmental benefits when considering the effect of substituting other polymers. When evaluating the results related to PHA/PHB co-production it has to be taken into account that the knowledge about impacts of plastic persistence in the environment is very low. The impact of persistent plastic might be of a larger impact than we can assess here. Similarly, extraction of polyphenols to be used for active packaging overall is an environmental benefit since they can potentially prolong shelf life of food and hence reduce food waste. Production and use of vine shoots or pomace as fillers in polymers is not always an environmental benefit compared to other uses of these residues. There are regional variations due to variations in the background system, mainly the energy system but also the offset for the products in the regions.

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3. Introduction

Task 6.1 in WP 6 concerns the application of the decision support approach issued from WP2 to selected conversion chains of agrowastes in order to identify and quantify environmental upscaling benefits. It seeks to identify the origin and magnitude of the potential environmental benefits of upscaling of the systems investigated in Task 6.2. Since the duration of the project has been too short to include learning related performance improvements of the upscaled system, modelling of (some of) the systems has been deployed in order to estimate the potential environmental performance improvements of the relatively mature system being investigated.

The technologies that have been pursued for upscaling in task 6.2 are primarily:

- PHA production (performed at the WP3 pilot scale platform by INNOVEN/UNIROMA)
- Solvent based extraction of polyphenols out of wine pomaces (scaled up and performed by FRAUNHOFER)
- Production of active packaging materials using polyphenols received after solvent based extraction (performed by FRAUNHOFER)
- Utilization of vine shoots (or extraction residues) for production of bio-composites with PHA (performed by INRAE)

PHA production already exists in pilot scale which is in operation at Isola della Scala (Verona, Italy) in the frame of Task 3.3. The scale seems to be adequate to foresee the impacts of a full scale plant and no further scale up is deemed necessary; however, technical problems persist in the improvement of the efficiency and the capacity of solid/liquid separation steps, which would increase the production capacity and the quality of produced polymer. The PHA production at this scale has been assessed in the publications Vega et al., 2019, Vega et al., 2020a, Ekman Nilsson, 202X. In relation to PHA-production two technology systems have been assessed; a biogas only scenario producing biogas and digestate, and a PHA-biogas scenario producing PHA, biogas and digestate. The multi-product output is included in the LCA through system expansion and biogas is valorized in a combined heat and power engine (CHP) substituting electricity (Vega et al., 2019) or electricity and heat produced by average technology in the region (Vega et al., 2020a) whereas PHA substitutes fossil plastic production (in Vega et al. (2020a) it is the average global thermoplastic production, whereas in Vega et al. (2019)) it substitutes PLA or PET). The assessment in Vega et al. (2019) is performed for two regions, one in Southern France and the other in Oregon, USA. Changing energy systems are taken into account via multiple dynamic energy provision scenarios. The assessment in Vega et al. (2020a) considers Bavaria and Veneto.

The pilot scale solvent based extraction of polyphenols has progressed well at Fraunhofer with acetone. For the LCA studies, data on the two extraction methods, solvent extraction (SE – with Acetone) and pressurized liquid extraction (PLE – with ethanol) was collected from the technology developers. An overview of the process steps can be seen in Vega et al. (2020b). The model was scaled up using data from project partners and implemented using SuperPro Designer, with industrial equipment and scale, without altering key parameters such as yield, or solvent to dry weight (DW) ratios. Pilot scale results from Fraunhofer show that model upscaling of lab-scale from UNIBO and RISE are representative both in terms of yields, consumptions etc. The LCA studies based on these upscaled processes are therefore also representative. However, extraction with acetone will be a problem both from a health and a safety perspective in the extraction plant as well as from the application side where polyphenol extracted with acetone may be prohibited in food packaging applications. Therefore, it is probable that ethanol-water

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extraction will be the preferred pathway in industrial scale even if less environmentally friendly. Production of active packaging has been demonstrated at Fraunhofer, as well as at ITRI. It seems that polyphenols adhere well to PET and PLA but a little less on Polypropylene. LCA of the polyphenol production has been assessed in the publication Vega et al, 2020b. In Ekman Nilsson et al 202x polyphenols were assumed to substitute ascorbic acid since their performance in active packaging application is comparable. The potential environmental benefits of using polyphenols as active packaging has not been fully investigated due to insufficient knowledge about the potential increase in shelf life and how that would potentially reduce food waste and production of food. However, some preliminary calculations have been prepared in this deliverable.

Production of bio-composites using PHA and vine shoots has been investigated at INRAE/University of Montpellier and LCA studies were performed to compare composites with different polymers and different weight fractions of vine shoot fillers. The LCA compared first rigid virgin polymer trays made out of PHBV, PP, and PLA and then the same polymer matrices with the addition of vine shoots filler material. The assessment was performed from cradle to grave with a cut-off system. These results are presented in David et al., 2020.

The manuscript in preparation (Ekman Nilsson et al., 202x) provides an overall assessment of biorefineries with different feedstocks and different output products and covers all the technologies mentioned above. The study has a regional perspective and the environmental performance is assessed in several European regions (Veneto, Bavaria, Languedoc-Roussillon and Skåne) and Oregon in the US. Biorefinery concepts were modelled for the raw material/feedstock availability in each particular region. For this study, the technologies were analyzed in a modular way, using the results generated on the following biotechnologies: polyphenol extraction, AD and AD+PHA, and filler, respectively in addition to many synergistic combinations, see figure 1. Overall, Biogas average utilization has been assumed which includes: Valorisation in CHP, electricity produced substitutes average energy mix of the country, heat is utilized for District Heating (DH) only for DE and SE. Upgraded to biomethane of natural gas grade (IT, DE, SE), for utilization in transport (SE). PHA and biocomposites- substitutes average global thermoplastic production 1:1, and 0.3:1 ratio as granules, respectively. Polyphenols – substitute ascorbic acid, ratio of 1:1. Digestate - substitutes manure and chemical fertilizers and their application. Field emissions are not included. Reduced emissions of methane resulting from reducing storage of manure are included. This results in 16 mini-LCAs per region assessing the treatment of 1 ton of agricultural residues (100% the same residue), though the residues considered were limited to animal manures, straw, wine pomace and vine shoots. Regional feedstock is then used as the functional unit with guidance from the mini-LCAs in order to choose the best performing feedstock and biotechnology pairings for each region. This results in a given regional system utilizing the overall most environmentally beneficial mix of technology-feedstock pairings as possible.

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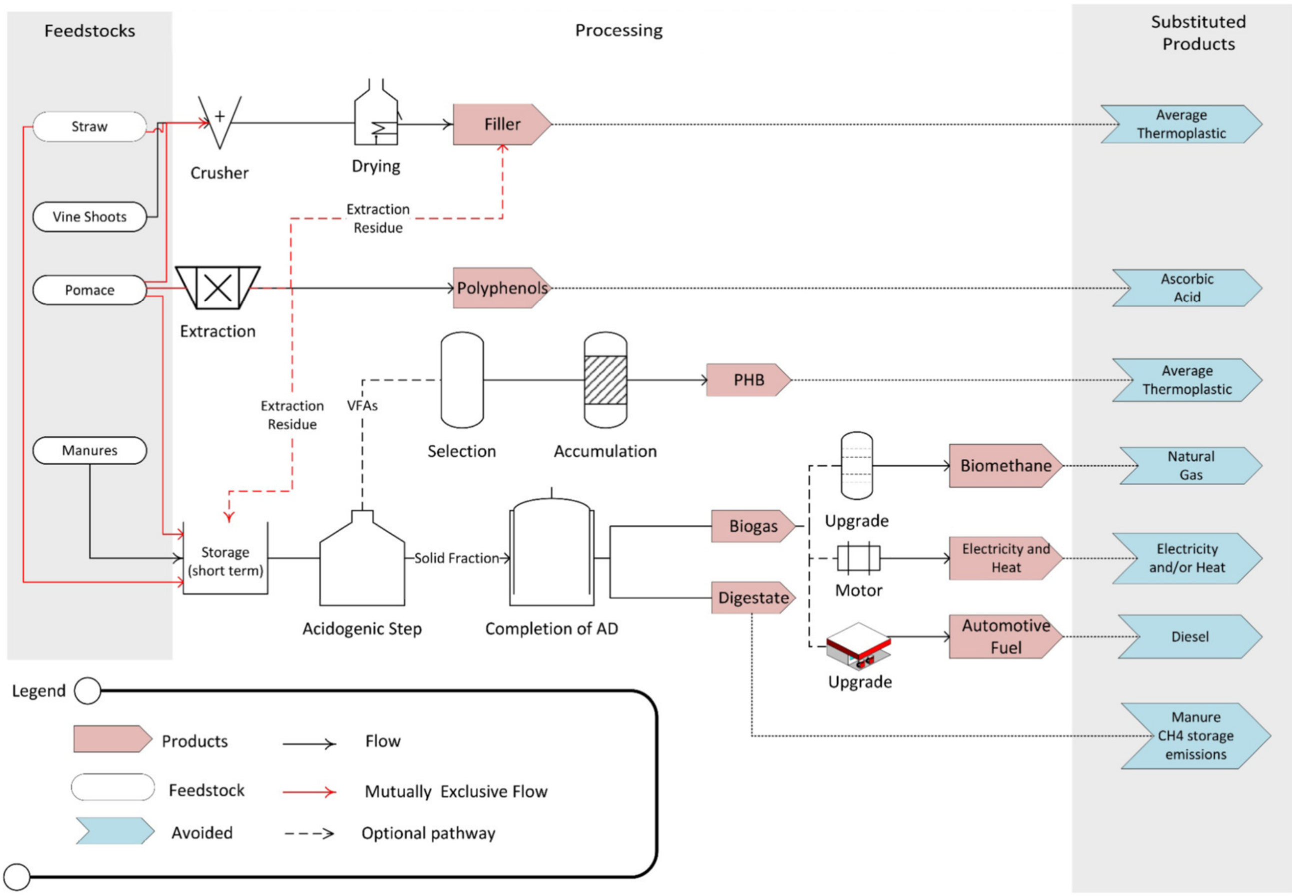

Figure 1: Technology to feedstock compatibility, showing possible combinations for the biotechnologies assessed (taken from Nilsson et al. 202x)

Overall, the OpenLCA software (GreenDelta, 2019) was used for the LCAs, along with the Ecoinvent v3 database (Wernet et al., 2016). The ReCiPE Hierarchist (H) (Huijbregts et al., 2017), method was used for impact characterization. All impact categories were included in the assessments and analyzed at midpoint and in some cases at endpoint. Territorial Metabolism-Life Cycle Assessment (TM-LCA) framework was applied in Vega et al. (2019), Vega et al. (2020a) and Nilsson et al (202x).

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4. Results

4.1. Results regarding PHA production

There are overall three different LCAs on anaerobic digestion (AD) with combined production of biogas and PHA. All of these are based on the pilot scale plants in Veneto, but modelled for varying geographical regions. The first study that was carried out compared PHA and biogas production in Languedoc Roussillon (FR) or Oregon (US) and did take into account dynamic development of the electricity production in those regions, i.e. considering what the electricity production from biogas in CHP would substitute in the future, but did not take potential use/substitution of heat into consideration. It considered that PHA substituted either PET or PLA. In this study it is evident that co-production of PHA is environmentally beneficial (see figure 2), no matter if it substitutes PET or PLA, and will be even more so in the future. Whereas biogas production only substitutes electricity production the co-production of PHA additionally substitutes other polymers leading to an increased saving as shown in figure 2. The figure only shows Global warming potential, but it was equally beneficial in relation to almost all other environmental impact categories, see the appendix for details.

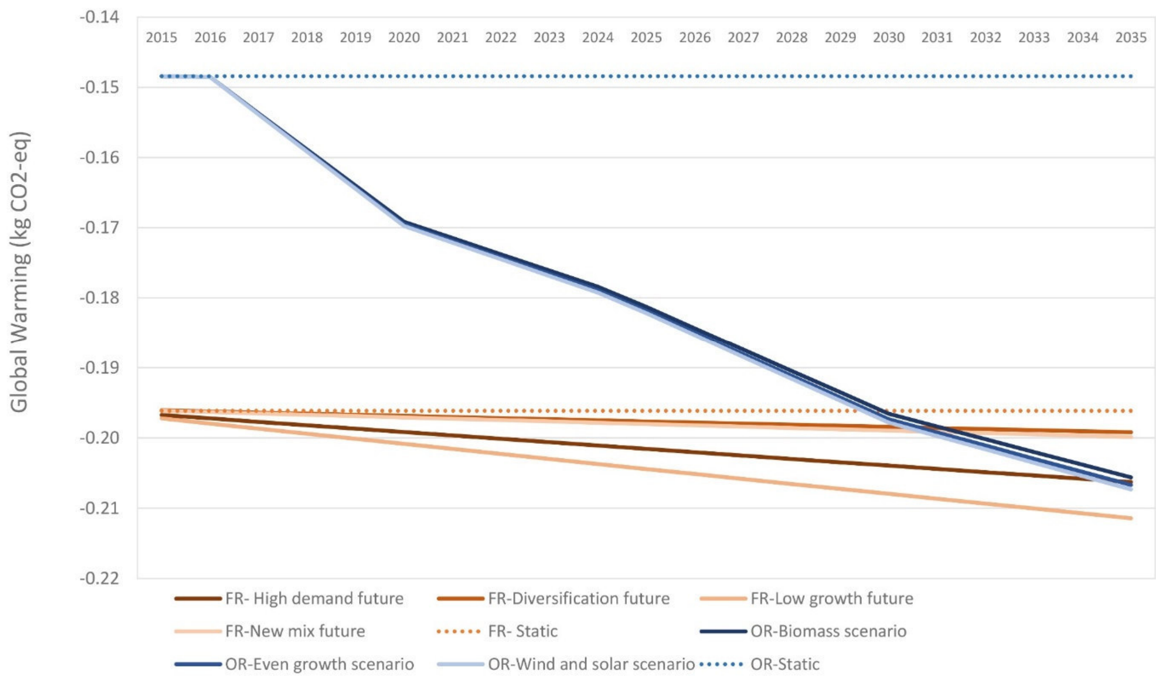

Figure 2. Yearly difference of Global Warming potential impacts i.e. PHA-biogas minus Biogas-only scenarios. Figure reflects the evolution of the energy mixes in the two locations. Negative values mean PHA-biogas has higher savings than Biogas-only. (Vega et al., 2019)

In the second study, AD with or without PHA (or actually PHB) co-production was assessed in Bavaria and Veneto considering the regional feedstocks, as well as at two different scales (200 kW and 1 MW). Additionally, the use of AD booster technology was assessed. The environmental impact (and

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economic) potential of each technology when scaled up to the regional level, meaning that it considers all of the region's unique sustainably available feedstock, was assessed.

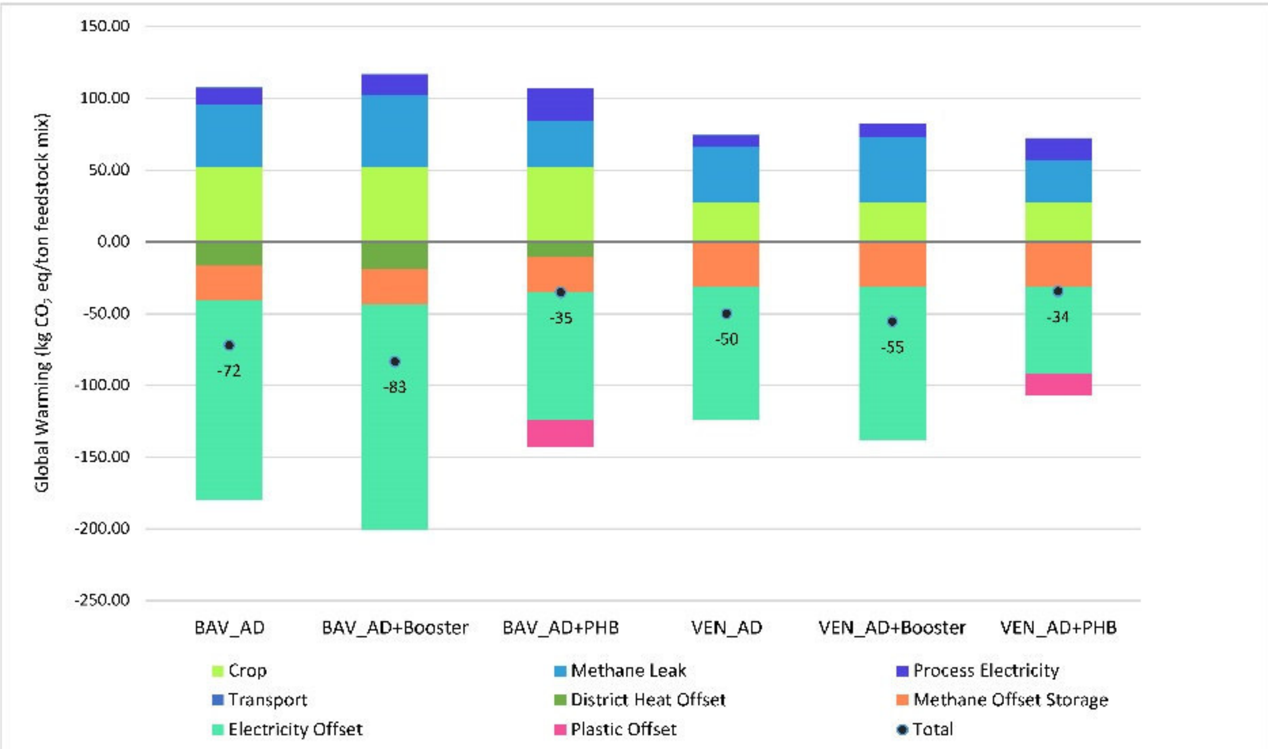

Figure3 GWP contribution per ton of feedstock mix for the two regions, BAV for Bavaria and VEN for Veneto, for the three technology options i.e. AD, AD+Booster and AD+PHB. Source: (Vega et al., 2020a). "Crop" is primary produce e.g. the Maize silage added in order to improve digestion.

The results are not as clear as in the first study. It is undoubtedly an environmental benefit to produce PHA/PHB. However, focusing on biogas production, especially using booster technology will bring even larger environmental benefits. The improvement modelled in this case, which is for energy efficiency of the PHA-biogas technology overall, is not the only factor that might improve for this technology in the future. Yield improvements for the polymer production might indeed be a more important factor of improvement. However, correct determination of a future yields for the technology are more difficult to determine. In this case it would be advisable that future research includes possible yield improvements which could be included as scenario ranges, which test extremes.

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Preference for the technology scenario producing the most energy was shown for all regions and scales, while producing bioplastic was less preferable since the value of the produced bioplastic was not great enough to offset the resultant reduction in energy production. Assessing alternatives in a regional context provided valuable information about the influence of different types of feedstock on environmental performance.

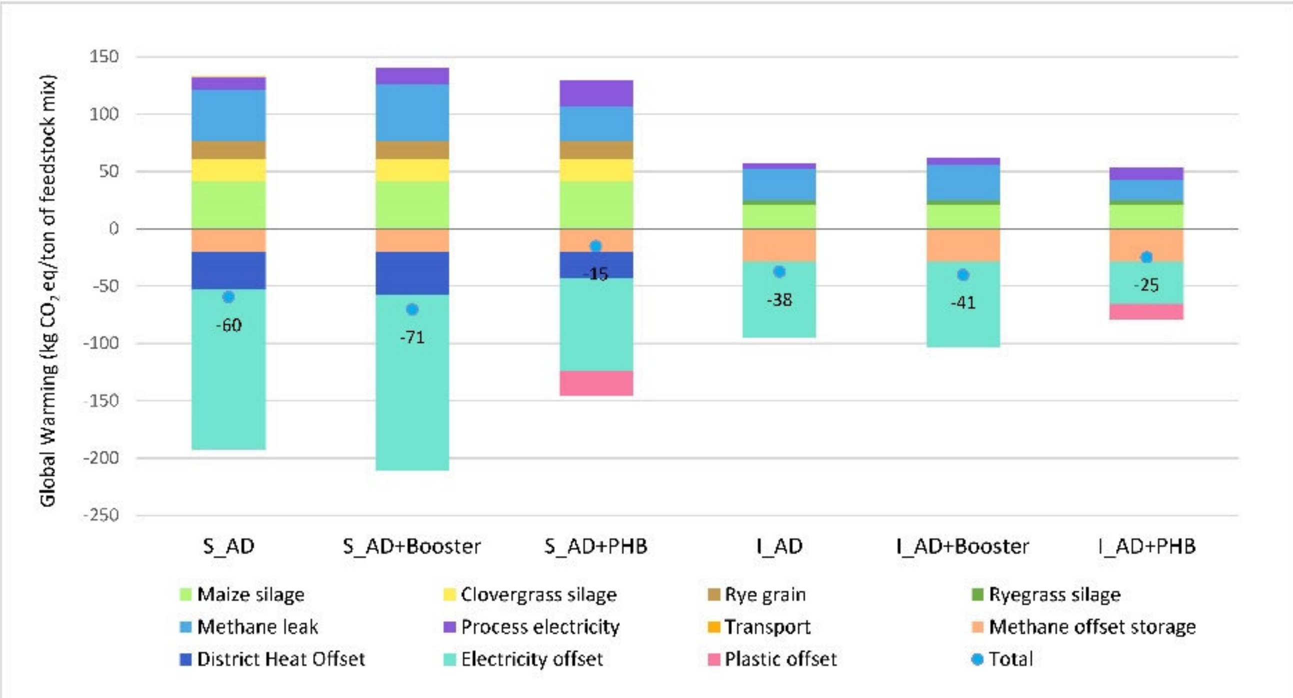

Figure 4 Global warming potential results for the small scale (200 kW) and Industrial scale (1000 kW) cases, per ton of feedstock, as well as contribution to GW by each stage. Scenarios are named as S for small scale and I for industrial scale followed by each technology scenario (AD, AD+Booster, AD+PHB). Source: (Vega et al., 2020a).

In the third study comparing 5 regions and different manure feedstock the picture was more or less the same as in the second. Focus on production of biogas was environmentally beneficial compared to co-production of biogas and PHA/PHB, although the production of PHA/PHB in it selfwere environmentally beneficial.

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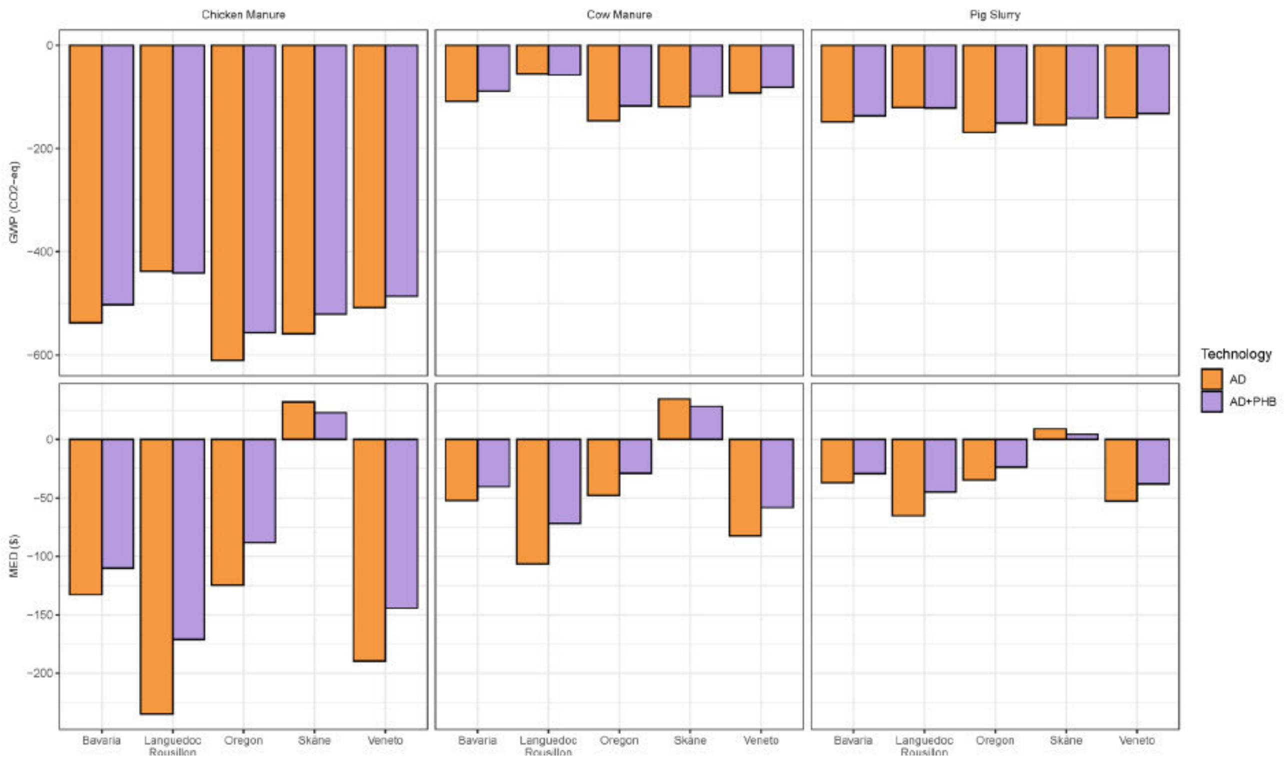

Figure 5: GWP impacts and MED (Monetised environmental damage) for manure feedstock-region-technology pairings Ekman Nilsson et al (202x).

4.2. Results regarding polyphenols extraction

Please see an overview of the production process in Vega et al. (2020b). It is shown that Solvent Extraction with acetone (SE) performs better (in term of having less environmental impact) than Pressurised liquid extraction (PLE) using a mix of ethanol and water. This is primarily due to a lower solvent to DW ratio and a less expensive processing setup. Interpreting the results, energy used for cooling and heating for distillation as well as energy for compressing the system dominate the CO_2 burden . However, through process optimization it is possible to drastically reduce some impacts (Carbon foot prints) that were large in the laboratory scale, as for example the impact from the spray dryer for the SE options, by adding a concentration (filtration) step before the drying. On the other hand, it is possible to see that adding a drying step for the pomace in option SE-2, does not pay off in comparison to not drying in SE-5, as the dryer plus distillation heating and cooling, are on the same range of impact as just distillation heating and cooling in SE-5. The overall GWP is lower than lab-scale for all options due to the reduction in solvent use and addition of extraction steps. In the overall LCA assessment, added acetone or ethanol weigh more than added heat or electricity, with acetone being two times more burdensome than ethanol. Nevertheless, the use of solvent in the PLE options is high enough that even though ethanol is less burdensome the total impact outweighs the acetone use in the SE options. But as mentioned, it is probable that an industrial scale extraction would use ethanol rather than acetone for health and safety reasons.

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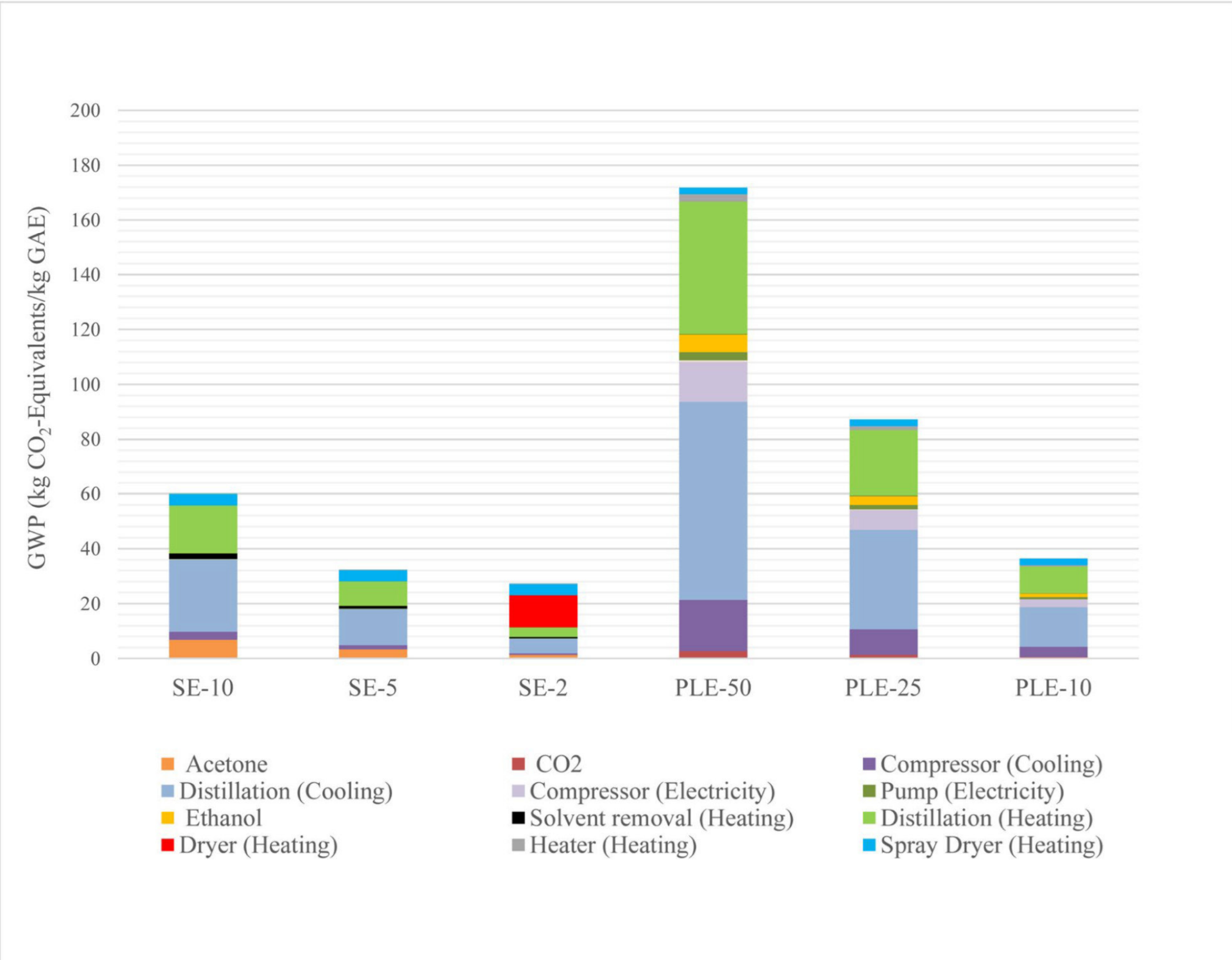

Figure 6: Global warming potential for scenarios tested in kg of CO2-equivalents. Contribution per processing step, cutoff 1% of overall impact. SE is solvent extraction with acetone, while PLE is pressurized liquid extraction using ethanol/water. The number at the end of each scenario indicates the solvent to DW ratio for the extraction process. Source: Vega et al., 2020b

Polyphenols in active packaging:

There are few studies that examine the relation between the shelf life of a product and how much of the product ends up as waste. One study that has aimed to examine how much of common food products that are wasted and if a packaging with higher environmental impact can motivate less food loss is Heller et al. (2018). In this deliverable we do very rough calculations to get an idea, if the increased environmental impact caused by coating a package with polyphenols extracted from wine pomace can be motivated by reduced food waste. The calculations are largely based on assumptions, because we cannot tell how much the waste is reduced if shelf life is increased by active packaging neither through project

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results nor literature. The active packaging is assumed to be used only for meat products. In the calculations we only considered waste that occurs in retail, not in households.

Heller et al. (2018) estimated losses of meat products in retail in the US to be between 3.5%-4.4%. In our calculations we assumed food losses in retail to be reduced by 50% by the use of active packaging that increases shelf life.

In these preliminary LCA calculations we have assumed that the only difference between the active packaging and the conventional packaging is the polyphenol coating. According to the experiments performed by Fraunhofer, the coating is 11.8 µm thick and contains 0.00259 mg GAE/cm². Meat is assumed to be packed in 500g portions each with an area of 20*15 cm. Only the polyphenols are included even though the coating also has other contents. Data for the extraction of polyphenols is taken from Vega et al. (2020b).

Table 1: Comparing the impact of food loss with the impact of producing polyphenols.

Meat type[1]	Impact of losses (4%) (kg CO2-eq)	Impact of losses (2%) (kg CO2-eq)	Impact of losses (0.7%) (kg CO2-eq)	Impact of losses (0.35%) (kg CO2-eq)	Impact polyphenol coating (kg CO2-eq)[2]
Beef	1.12	0.56	0.196	0.098	$3.89 \cdot 10^{-5}$ - $9.32 \cdot 10^{-5}$
Pork	0.164	0.082	0.0287	0.01435	$3.89 \cdot 10^{-5}$ - $9.32 \cdot 10^{-5}$
Chicken	0.104	0.052	0.0182	0.0091	$3.89 \cdot 10^{-5}$ - $9.32 \cdot 10^{-5}$

[1]Data on meat production is taken from RISE Climate Database (2020). Beef 28 kg CO2-eq/kg, pork 4.2 kg CO2-eq/kg and chicken 2.8 kg CO2-eq/kg. [2]Variation between calculated best and worst case for solvent extraction of polyphenols from grape pomace.

As can be seen in the table, even at the lowest level of food waste and worst-case extraction process, active packaging with polyphenols have a great potential to improve environmental performance in the food chain. However, these preliminary calculations should only be seen as indications and should be completed with actual figures including additional environmental impact categories.

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4.3. Results regarding biocomposites

This study showed that bioplastics are currently less eco-friendly than PP, which would be a potential polymer to mix in fillers. This is in part due to PHA having a higher density than PP resulting in a higher weight of the PHA trays compared the to PP trays. But also to a large extent due to the fact that LCA does not account for, in existing tools, effects of microplastic accumulation resulting from fossil based plastic and that bioplastic technologies are still under development with low tonnage. This study also demonstrated the environmental interest of the development of biocomposites by the incorporation of Vine shoot (ViSh) particles. The minimal filler content of interest depended on the matrices and the impact categories. Concerning global warming, composite trays had less impact than virgin plastic trays from 5 vol% for PHBV or PLA and from 20 vol% for PP, as shown in figure 7. It can therefore be concluded that use of fillers is beneficial if higher than 5 vol%, since the primary objective is to use them as fillers in PHA/PHBV. Concerning PHBV, the only biodegradable polymer in natural conditions in this study, the price and the impact on global warming are reduced by 25% and 20% respectively when 30 vol% of ViSh are added. Figure 7 sums up the findings related to fillers.

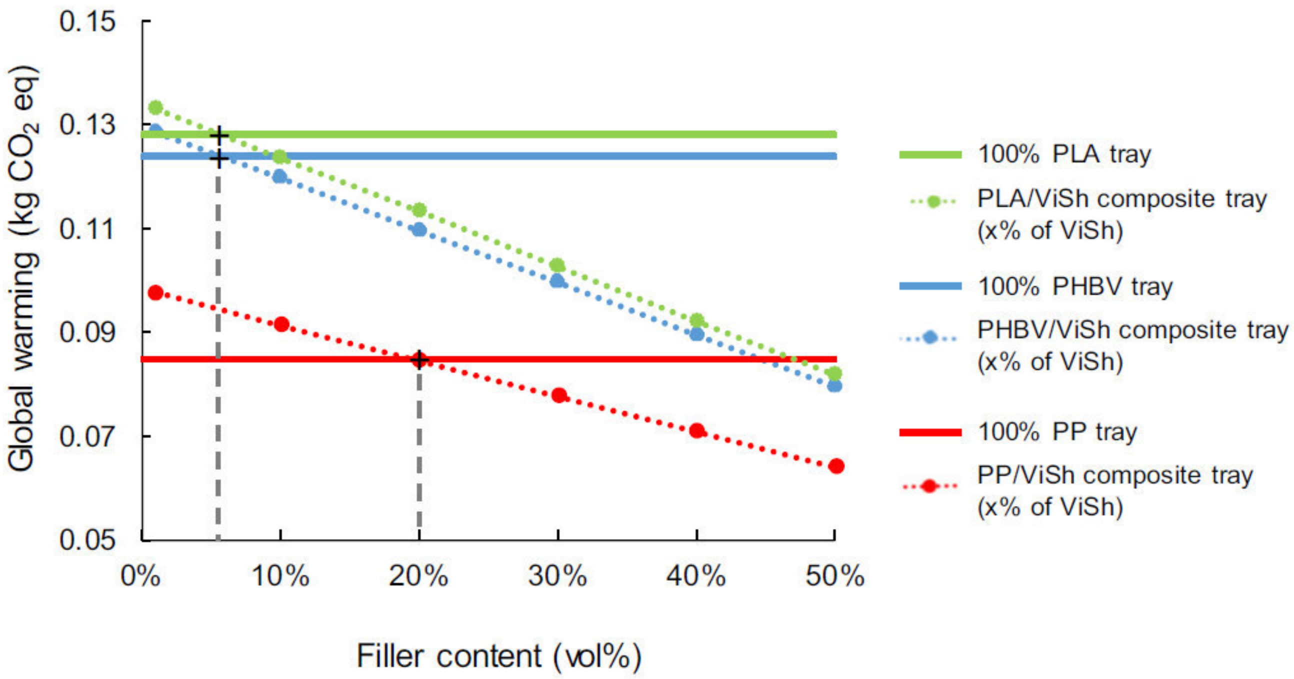

Figure 7: Global warming impacts as a result of the filler content. (Taken from David et al., 2020) (ViSh = Vine shoots)

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4.4. Overall results on biorefineries

As illustrated in figure 8 there is no single answer to what is the environmentally preferable use of agricultural waste/ feedstock. It depends to a large extent on the energy system that is substituted by the biogas, which, as shown in Vega et al. (2019), is ever moving towards more renewable energy sources. This dynamic change will with time reduce the benefits of biogas. The results were shown for liquid residue feedstock in 4.1 and the figure 8 shows the results for solid residue feedstock. Looking only at straw and Vine shoots they are generally a benefit to use both as feedstock for AD and as filler. Sweden is a special case due to due differences in the electricity mix with a larger share of low carbon energy such as nuclear and hydro. Generally, the same is true for pomace. However, the extraction of polyphenols seems not only to be positive in contrast to the preliminary calculation in chapter 4.3. This is because in the study shown in fig. 8 only a substitution of ascorbic acid was considered – not the potentially reduced food loss.

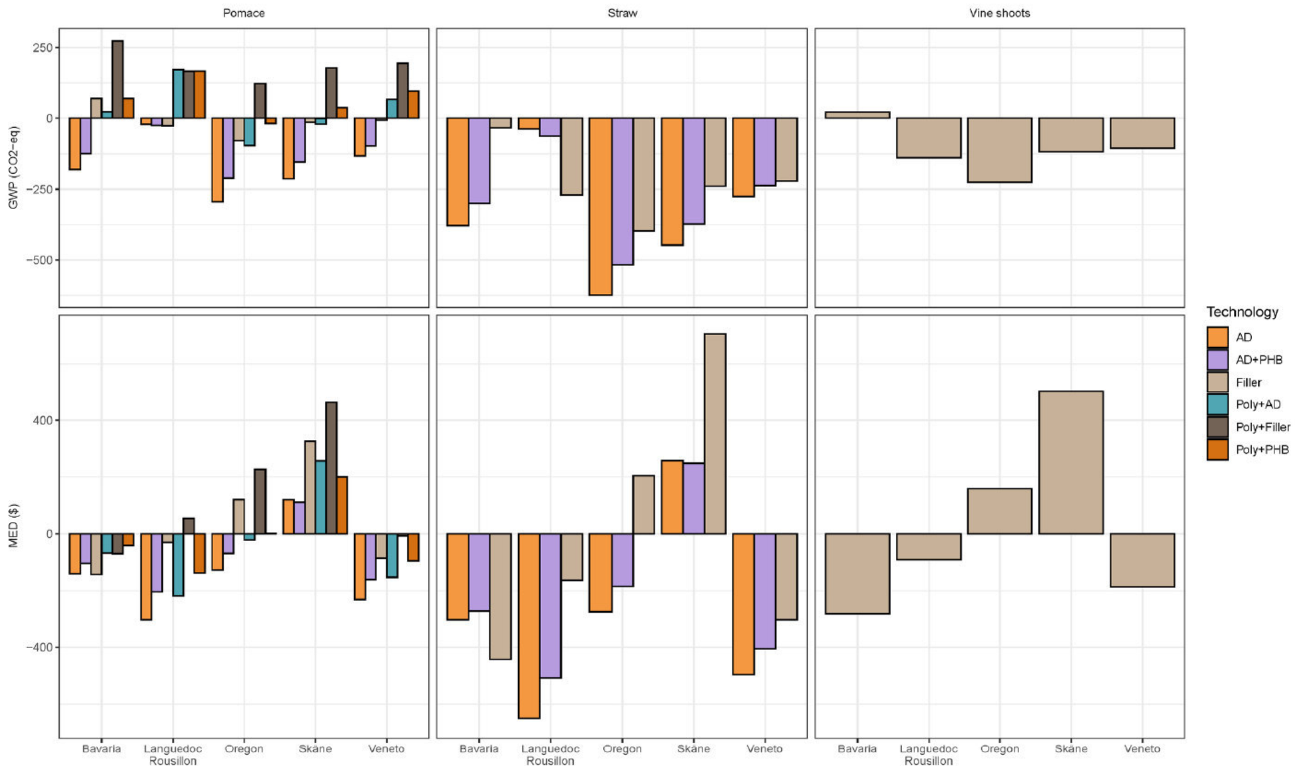

Figure 8: Global warming potential (GWP) impacts and monetized environmental damages (MED) for solid residue feedstock-region-technology pairings (EkmanNilsson et al, 202x)

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5. Conclusions

Overall, it is evident that the different technologies that are considered up-scaled in NoAW all have their benefits, even though some are more environmentally beneficial than others. Table 2 below summarises nicely which technologies are beneficial from an environmental perspective taking into account the energy systems in the region and the available feedstock. It can be seen that the low carbon energy system of Sweden makes it difficult to obtain high environmental benefits, which may give an indication of how it can look for other regions in the future. Nonetheless, the environmental benefits can be seen for all technologies. However, there are contradictions between GWP and Monetized environmental impacts. Regional variations are due to variations in the background system, mainly the energy system but also the offset for the products in the regions. For filler and PHB also the end-of-life is a contributing parameter.

When evaluating the results related to PHA/PHB co-production it has to be taken into account that the knowledge about impacts of plastic persistence in the environment is very low. The impact of persistent plastic might be of a larger impact than we can assess here.

It may be quite difficult to get an overview of whether the options are beneficial or not which is why Ekman Nilsson et al (202x) made a summary table that is shown below.

Table 2: Impact of technology value chain implementation for Bavaria (DE), Veneto (IT), Languedoc Roussillon (FR), Skåne (SE), and Oregon (US) for both global warming potential (GWP) and monetized environmental damages (MED) (Ekman Nilsson et al., 202x)

	DE MED	DE GWP	IT MED	IT GWP	FR MED	FR GWP	SE MED	SE GWP	US MED	US GWP
AD	Induced savings	Induced savings	Induced savings	Induced savings	Induced savings	Induced savings	induced impact	Induced savings	Induced savings	Induced savings
AD+PHB	Induced savings	Induced savings	Induced savings	Induced savings	Induced savings	Induced savings	induced impact	Induced savings	Induced savings	Induced savings
Polyphenol -extraction	na	na	Induced savings	induced impact	Induced savings	induced impact	na	na	na	na
Filler	Induced savings	induced impact	Induced savings	Induced savings	Induced savings	Induced savings	induced impact	Induced savings	induced impact	Induced savings

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6. References

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7. Partners involved in the work

Technical University of Denmark

Research Institute of Sweden

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8. FAIR Data management

Data set related to Vega et al. (2019) is published as https://doi.org/10.15454/ABCM1W

Data set related to Vega et al., 2020a is published as https://doi.org/10.15454/IXFDKI

Data set related to Vega et al., 2020b is published as https://doi.org/10.15454/LZCMKQ

Data set related to David et al., 2020 is published as https://doi.org/10.15454/RLO20C

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9. Annexes

The referred papers Vega et al., 2019, Vega et al., 2020a, Vega et al, 2020b and David et al, 2020 are appended. Ekman Nilsson et al., 202x is not yet included since it is not published yet.

sustainability

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Article

Maximizing Environmental Impact Savings Potential through Innovative Biorefinery Alternatives: An Application of the TM-LCA Framework for Regional Scale Impact Assessment

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Abstract: In order to compare the maximum potential environmental impact savings that may result from the implementation of innovative biorefinery alternatives at a regional scale, the Territorial Metabolism-Life Cycle Assessment (TM-LCA) framework is implemented. With the goal of examining environmental impacts arising from technology-to-region (territory) compatibility, the framework is applied to two biorefinery alternatives, treating a mixture of cow manure and grape marc. The biorefineries produce either biogas alone or biogas and polyhydroxyalkanoates (PHA), a naturally occurring polymer. The production of PHA substitutes either polyethylene terephthalate (PET) or biosourced polylactide (PLA) production. The assessment is performed for two regions, one in Southern France and the other in Oregon, USA. Changing energy systems are taken into account via multiple dynamic energy provision scenarios. Territorial scale impacts are quantified using both LCA midpoint impact categories and single score indicators derived through multi-criteria decision assessment (MCDA). It is determined that in all probable future scenarios, a biorefinery with PHA-biogas co-production is preferable to a biorefinery only producing biogas. The TM-LCA framework facilitates the capture of technology and regionally specific impacts, such as impacts caused by local energy provision and potential impacts due to limitations in the availability of the defined feedstock leading to additional transport.

Keywords: biorefinery; territorial metabolism; life cycle assessment; biogas; multi-criteria decision assessment; bioplastic; polyhydroxyalkanoates; agricultural residues

1. Introduction

Life cycle assessment (LCA) is a tool designed to quantify the environmental impact potential of products and services [1]. Recent advances in the field of LCA, such as the inclusion of temporal dynamism [2] and the coupling of LCA to urban metabolism [3] increase the applicability of the LCA methodology. Dynamism in LCA allows for the quantification of impacts while taking into consideration changing background and foreground systems, e.g., amounts of renewable and fossil energy sources in the electrical energy mix of a specific location in the background, and improvement to processing technologies in the foreground. On the other hand, coupling urban metabolism to LCA allows for large-scale assessments that better predict large-scale consequences of implementing a change at regional scale. These advances are an especially important input that can help guide the transition into a sustainable bioeconomy, as they allow for prospective studies. LCA of production systems/technologies, such as various agricultural productions, e.g., wine, cereal, and meat, can benefit from applying some of the new developments, since the large inputs and outputs to these systems, most likely, will have great environmental implications when changes to the production are implemented.

By applying the TM-LCA framework, as used in this study, it is possible to assess said systems in the specific context of the region, i.e., taking into consideration the region's infrastructure, feedstock availability and accessibility, and the technical feasibility of technology implementation. Assessing large systems, as mentioned above, can be approached by defining the geographical boundaries in terms of a "producer territory" [4] so that the LCA can be applied for assessment of a delimited "territory", e.g., wine-producing areas, within a broadly defined region, e.g., Southern France. The producer territory is thus defined as the area of interaction between the aggregated producers and other systems within the region. The TM-LCA framework reduces data demand by aggregating individual areas of the production of, for example, a specific product, supply chain or waste treatment technology, while ignoring unchanging background systems, i.e., only changes to the region interacting with the producer territory are assessed. At the same time, representativeness is increased by merging local inventory data from individual producers with regional and nation-wide data in order to fill in data gaps. In this way, an environmental performance improvement in the territory, due to, e.g., the implementation of a new technology or new management technique, can be quantified in the non-contiguous production area and is reflected in the results for the region. When combined with dynamic and prospective LCA [2], this approach offers a comprehensive assessment that gives temporally and geographically resolved results. Moreover, it has the added utility of providing prospective insights that can more accurately support decision makers, production owners, and technology developers [4].

A point of departure for many LCAs is a static product system, where, for example, technology A might be assessed against technology B for the making of a product. The static nature of LCA is problematic when applied to products or systems with long service lives [5], due to inconsistencies in time horizons and changes in background systems [6,7]. Previous work has demonstrated the importance of incorporating various types of dynamism into LCA, as this can significantly affect the results of the study [6]. In this regard, it is possible to add dynamism to the various stages of the LCA in a consistent, systematic, and transparent manner, as outlined in [2] and shown in various other publications [7–9]. Following the TM-LCA framework, dynamism can be added in a consistent manner from the start, which provides added information regarding the sensitivity of the system to background changes. Real production systems are rarely static, and results based on static systems can sometimes exhibit rank reversal when compared to dynamic results [10]. Thus, basing future decisions on static LCAs can result in building significant error into the models and associated results. Adding dynamic aspects to LCAs can increase the analytical accuracy of results [11].

The added layers of information to the TM-LCA mean that the interpretation phase becomes more resource demanding. This can be eased by the use of extra tools, such as multi-criteria decision assessment (MCDA). Midpoint results for 18 different impact categories of an LCA are often difficult and time consuming to synthesize into clear and readily applicable decision support. When adding dynamism, this translates into temporally specific results for, e.g., each year of the time horizon, for each of the 18 impact categories. Out of the many MCDA methods that exist, one that has shown great capability in dealing with LCA results is Technique for Order of Preference by Similarity to Ideal Solution (TOPSIS) [12,13]. The output from TOPSIS is given in the form of a single score performance index, which is used to derive preference between the scenarios being assessed. By checking a multiple criteria decision support tool used with equal weightings for all midpoint impact categories, it is easy to realize and visualize burden shifting amongst the midpoint impact categories, when used in conjunction with a visual inspection of internally normalized results. The MCDA approach is considered preferable, as using carbon footprint alone has been shown to give potentially misleading results [14].

The present study's goal is to implement an assessment based on the TM-LCA approach [4] in order to provide a comparison of potential biorefinery choices for the treatment of agricultural residues. For the demonstration of TM-LCA, a biogas production scenario is compared to a scenario of combined biogas and Polyhydroxyalkanoates (PHA) production, which is currently being developed at pilot scale. Polyhydroxyalkanoates are naturally occurring polymers produced by a consortium of bacteria, which can feed on the volatile fatty acid (VFA) stream generated by the acidogenic phase of anaerobic digestion (AD) [15]. PHA, which is also found as polyhydroxybutyrate (PHB), can be used to produce biodegradable plastic products. In this case, PHB production substitutes the production of polyethylene terephthalate (PET) or polylactide (PLA). The two biorefinery scenarios are modeled with dynamics built into both foreground and background systems. In the foreground system, dynamics are included as a yearly decrease in the amount of energy consumption needed to produce PHA. In the background system, the electrical energy mix, hereafter referred to as energy mix or energy grid, of both locations is varied yearly for a period of 20 years with four possible provision mixes for Oregon, and five possible choices of provision for the energy mix futures of France. The scenarios are then tested at a territorial scale as described above, i.e., processing all the feedstock in the region in the two geographically dissimilar production territories, to observe the effects of regional differences on territorial performance. Since the use of global warming potential (GWP) as a single indicator has been shown to provide potentially misleading results [14], MCDA is applied in the interpretation phase to help ease the interpretation of results.

2. Materials and Methods

2.1. TM-LCA Framework Application

The application of the TM-LCA framework is described in general terms here. A point of departure for the application of the TM-LCA framework is the functional unit. The functional unit, the treatment of one ton of feedstock of specific composition, is treated by two different technology alternatives, described in more detail below. From here, the following steps are applied and described through the methodology:

(a) Alternative technology is defined.
(b) The producer territory is defined and limited to systems interacting with the technological options being assessed within a geographical region.
(c) Temporal dynamics are incorporated into the systems, e.g., in dynamic background electricity energy provision and technological efficiency improvement.
(d) The assessment is scaled to encompass the whole region so that all feedstock available that may fulfill the functional unit is treated by the technological alternatives being assessed. However, only changes in systems and in the region are assessed.

2.2. Goal and Scope

In order to implement the TM-LCA framework, two options for the treatment of agricultural residues were modelled and compared in two geographic locations, the Languedoc-Roussillon region in southeast France and the Willamette, Umpqua, Rogue, and Columbia valleys of Oregon State in the USA. Advancements in biogas technology make it possible to treat a plethora of agricultural residues, and recent innovation allows for the production of value-added products, in this case, the family of biopolymers known as polyhydroxyalkanoates (PHA). This innovative technology, which effectively creates a biogas platform for new biorefineries, is a contender to conventional biogas production where the only products are biogas and digestate. The proliferation of biogas plants makes this new addition to anaerobic digestion a highly transferable technology, which can be implemented wherever agricultural residues are available. Since biorefineries, in general, have a long service life (decades) and draw from large discontiguous areas, both territorial and dynamic aspects of this assessment are an advantage for decision makers considering biorefinery options for their region. However, it should be emphasized that the study only compares two different biorefinery types. It cannot be used to decide whether to increase the total use of residues for biorefineries.

Functional Unit

The basis for the comparison of the scenarios is the treatment of 1000 kg of feedstock. The feedstock is assumed to be agricultural residues of the following composition: 50% liquid cow manure, 15% solid cow manure, and 35% wine pomace or wine marc, hereafter used interchangeably. Feedstock characterization is based on laboratory tests performed onsite at an Italian biogas plant for the liquid and solid manure, while for wine pomace it is based on literature values. While other types of feedstock can be treated by the biorefineries being considered, the choice of feedstock was limited to the above in order to better appreciate the difference between biorefineries rather than differences arising from choice of feedstock. The feedstock physiochemical properties are presented in the supplementary information (SI).

2.3. Scenarios

Two baseline scenarios were assessed with the OpenLCA [16] software and the Ecoinvent 3.4 database [17]. The two alternative technological pathways possible for treating the functional unit are:

2.3.1. Biogas Only

Conventional biogas production was modelled as the anaerobic digestion step of biogas production, which produces biogas and digestate. The biogas was assumed to be burned in a combined heat and power (CHP) engine, producing electricity and heat based on the energy content of the biogas. Process energy consumption was calculated to be 7% of the electricity output, based on data received from an industrial scale biogas plant in Northern Italy, while the co-generated heat is assumed to be wasted. This is due to the geographical areas of implementation of the scenarios, where the excess heat is not used. Furthermore, adding the produced heat to this study would only change the magnitude of the savings from displaced energy production, and not the ranking of the scenarios, as seen in [18], as the magnitude of heat production is similar across scenarios. All other operational parameters were also based on the data acquired from the abovementioned biogas plant and are available in the supplementary information (SI).

Processing steps that are equal for both scenarios and emissions occurring therein, e.g., feedstock storage, animal housing and digestate storage, were excluded from the assessment, as they would result in no relative difference. Similarly, phosphorus fertilizer replacement was left out because the starting content of P is the same, and processing is not expected to change this. Adding replacement of P fertilizer to the assessment would only elucidate differences between digestate and mineral fertilizers, which is not the focus of this study.

2.3.2. Field Application of Digestate for All Scenarios

The field application of the digestate was modelled, and conventional ammonium nitrate fertilizer was assumed to be replaced. It is well known that digestates mineralize at a slower rate so that a share of the organic nitrogen present in digestate will be bound and will thereby not be available for crop uptake or emissions. Thus, an average mineral fertilizer equivalency value of 67.5%, calculated from a review of values that are commonly used in this type of assessment, was used for the substitution of mineral N fertilizer [19]. Emissions resulting from the field application of digestate were modeled based on the approach in [20], which applied the agronomic model Daisy [21] to estimate long-term emissions from different types of soils with different histories of management, i.e., high or low inputs of organic matter in the form of organic fertilizers, such as digestate and compost. As shown in this work, the crop's response to nutrient inputs is highly dependent on the previous fertilization history of the field. Emission factors (EFs) for high and low crop response after digestate application were taken from [22], which follows the same approach described by [20] and had soils and overall conditions which more or less match the soils in the geographical areas assessed here. For N_2O emissions, the Intergovernmental Panel on Climate Change (IPCC) methodology [23] and EFs were used. The sensitivity of N_2O EFs was tested in the sensitivity analysis due to the multiple models available for deriving EFs. The nutrient content of the digestates, as well as emission factors for all N-related emissions, for digestates and mineral N fertilizer are presented in Supplementary Tables S1, S2 and S4.

2.3.3. PHA-Biogas

The second scenario represents a tweaking to the AD process, where AD is split so that the VFA production that occurs during the first days of digestion is diverted and used to produce and feed biomass capable of producing PHA. Operational data from a PHA-producing pilot plant run by Innoven Srl were obtained and used to create an industrial scale model of PHA production. The co-production of biogas and PHA is executed, albeit with a lower biogas yield. Just as above, digestate continues to be produced and replaces mineral N fertilizer. Additionally, the extraction of polyhydroxybutyrate (PHB), a polymer in the family of polyhydroxyalkanoates, i.e., PHAs, is included as the addition of process energy consumption for the extraction, and hydrogen peroxide is included as an extraction agent. All other model parameters are equal to the biogas scenario.

PHA production is here assumed to be 100% PHB and replaces the production of petroleum or bio-based polymers, referred to as the replacement polymers (RP). In the first run of the model, PHB replaces PET at the factory gate, with a replacement ratio of 0.93:1 PHB to PET. In terms of material properties, several performance indices (PI) based on yield strength (σ), tensile strength, and density (ρ) were used to derive the replacement ratios (RR) (Equation (1)). The ratio of replacement is tested in the sensitivity analysis so as to represent different applications of the polymer more accurately. The choice of polymer substitution is also tested; since PHA is a bio-sourced biopolymer, a sub-scenario with replacement of biobased polylactide (PLA) is also presented. The RR is 0.64 for PHB substitution of PLA, based on Equation (1).

$$RR = \frac{PI_{PHB}}{PI_{RP}}, \text{ and } PI = \frac{\sigma}{\rho} \tag{1}$$

Equation (1) Polymer replacement ratio, where RR = replacement ratio, PI = performance index, σ = yield strength, RP = replacement polymer and ρ = density.

The addition of PHA production in this scenario is not burden-free, inducing impacts from energy consumption and via the production of the extraction agent. However, due to missing data from the pilot plant, the additional energy consumption was calculated using the process design software Superpro Designer® [24]. This yields an additional 7 kwh/functional unit (FU). It was assumed that process energy consumption for PHA could improve over time, so a 1% decrease in energy demand per year for PHA production was modeled for the assessed period. This represents the maturation of PHA extraction technology, which is a likely scenario as the implementation of PHA extraction in biorefineries becomes more widespread and further optimization of the technology takes place. This efficiency improvement rate is tested in the sensitivity analysis to explore the possibility of faster and slower improvements to the process. Key parameters for the production of PHB are presented in Supplementary Table S3.

2.3.4. System Boundaries

The system boundary of the two scenarios extends from when the feedstock enters AD to the application of digestate onto the field (see Figure 1). End of life was not included in the assessment, as the LCA methodology lacks an appropriate characterization of the effects from plastic degradation in the environment, such as microplastic formation and the production of methane among other decomposition gases [25,26].

Applying a dynamic approach, all background and foreground processes were modified so that the two geographical areas are accurately represented with likely different future energy production scenarios in accordance with the national and state-specific energy legislations and policies.

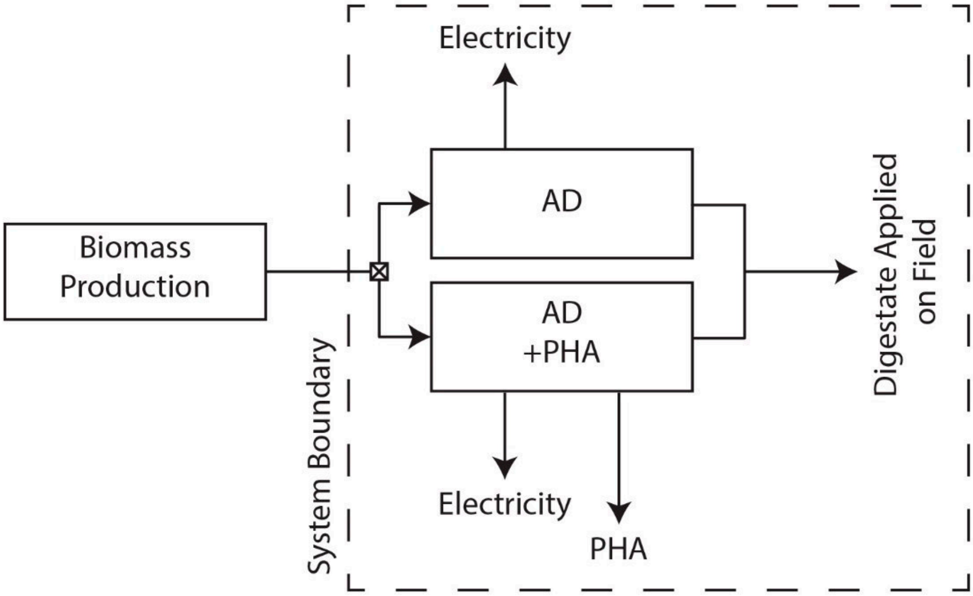

Figure 1. System boundary definition.

2.4. Dynamics

Dynamic inventories of the electricity mix for the two locations, modelled for a period of 20 years from 2015–2035, were used in the analysis. Four different dynamic energy futures, developed by the French government, with yearly shifting percentages of contributing sources of energy (Figure 2), were used for all electricity provision in the scenarios for Languedoc-Roussillon [27]. Likewise, three different dynamic energy futures were developed based on the legislation for Oregon State (Figure 3), which regulates the share of renewables in Oregon's future energy grid [28]. Qualifying renewables, i.e., renewable energy sources accepted by Oregon legislation on renewables, were introduced in varying amounts. Thus, (1) a scenario where biomass was increased more than other qualifying renewables, (2) a scenario where wind and solar were increased more than other qualifying renewables, and (3) a scenario where all qualifying renewables were increased evenly were developed. Static electricity mix scenarios were also included for both locations.

To maintain consistency in the foreground and background systems, the electricity provision component of all Ecoinvent processes used in the assessment was exchanged with the dynamic mixes developed. This included the electricity for fertilizer production, conventional polymer production, and the electricity replaced in the grid. This use of the local grid mix in the commodity production may not be a 100% accurate representation of a market reaction for the background systems, but it is deemed a better representation than the static processes. Further discussion on this subject can be found in Section 4.

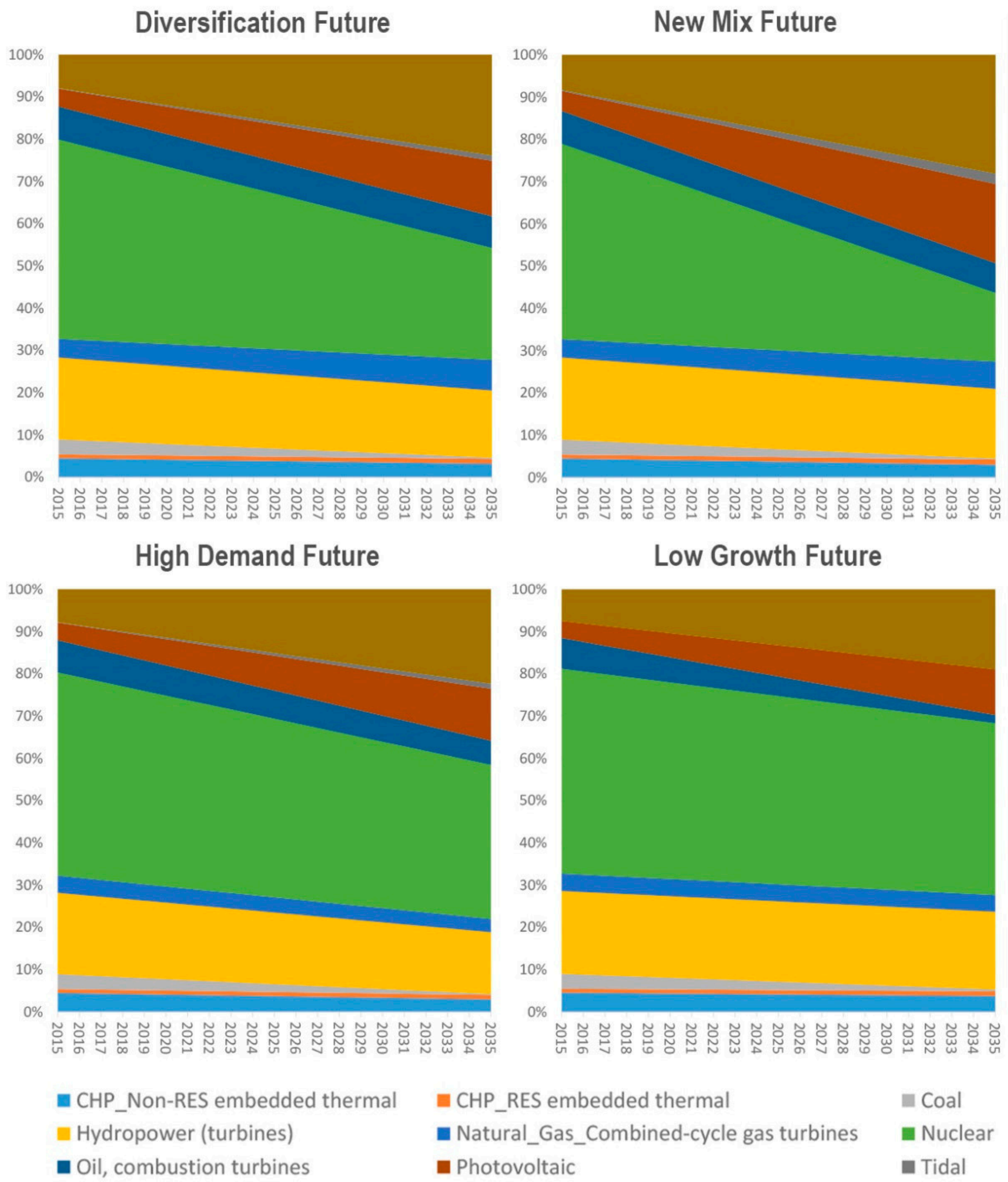

Figure 2. Evolution of the French electricity grid based on future scenarios defined by [27].

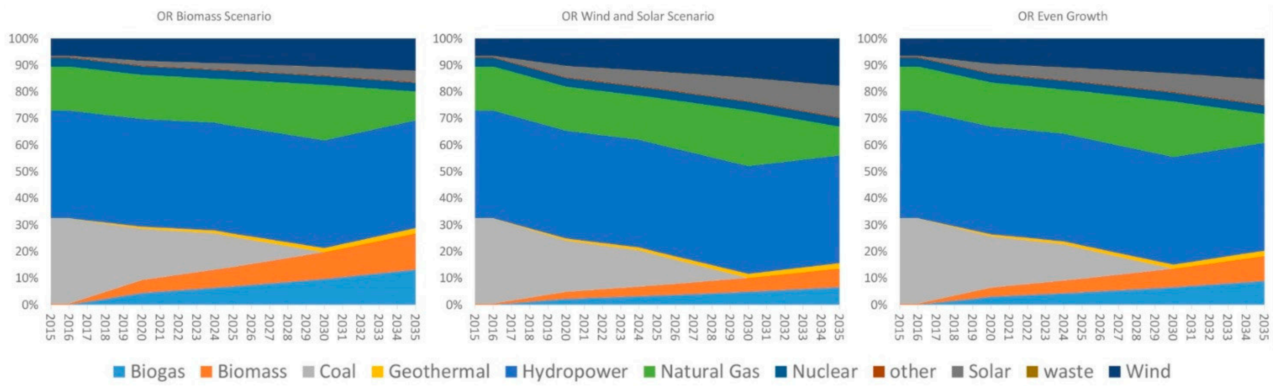

Figure 3. Evolution of the Oregon electricity grid based on three possible future scenarios for the fulfillment of legal requirements for decommissioning fossil-based production facilities.

PHA Process Energy Consumption

PHA production, which has been around since the 1980s, is already practiced at industrial level with first generation feedstock such as sugars from corn and sugarcane. Plants already exist with capacities ranging from 2000 to 50,000 tons of annual production [29]. Furthermore, PHA production has been introduced to the waste water treatment sector [30,31] and is also possible from second generation biomass. Due to important experience in the market with regards to PHA production, the PHA production for second generation biomass, as in the present study, will likely attain vast improvements in the future, eventually reaching a maturity level comparable to current industrial PHA production. To reflect this, dynamics in the PHA inventory were included in terms of electricity consumption (i.e., energy efficiency), in addition to the dynamic electricity provision. Hence, while PHA production was modelled starting as 7 kwh/FU more burdensome than the biogas-only scenario, thereafter the process was modelled as becoming more energy efficient, improving by 1% annually for the 20-year period, based on similar technology learning curves [32]. This improvement rate was also tested in terms of influence on total impacts (see Section 2.7).

2.5. Implementation of Territorial Scale Assessment

In order to assess the implications of implementing PHA technology at a territorial scale, the two study regions, in France and Oregon respectively, were analyzed regarding ability to provide feedstock for application in the two assessed biorefinery scenarios, i.e., impacts arising from treating all feedstock available in the region by biogas-only or combined PHA-biogas. The territories were defined as the interacting areas of residue production and the treatment plants. However, as defined in the TM-LCA method [4], only the areas undergoing change are included in the assessment. In this case, the change is an average change reflected in the residue treatment centers. Therefore, it is not expected that this change will affect the production of the residues in any way, ergo feedstock producers are left out of the assessment in terms of environmental impact. Likewise, transport from producers to treatment centers is not expected to change, as the volume of residues produced will not change as a consequence of implementing PHA technology. Where there is potential for transport that would deviate from the status quo, namely in the transport of grape marc which is the lighter of the two feedstock, impacts from transport were assessed (see 0, Sensitivity Analysis). These impacts were not included in the main results, as the induced impacts from transport would be equal in both the PHA-biogas and the biogas-only scenarios.

Feedstock Provision

Several assumptions were made in relation to determining the amounts of residue produced in each region for input into the regional scale assessment (Table 1). For wineries, it is assumed that grape marc is produced at a rate of 0.13 tons per ton of processed wine grapes [33]. It is further assumed that in France, where production data are reported in hectoliters of wine instead of mass of grapes at crush, 140 kg of grapes are used to produce 1 hectoliter of wine [34]. For feedstock coming from cattle, it is assumed that all waste comes from dairy cattle and that dairy cattle produce waste at a rate of 54.5 kg per head per day [35].

Due to the relative scale of wine production and the cattle industry in Oregon, the production capacity of the biorefinery systems in Oregon is limited by the production of grape marc, assuming that the co-digestion of cow waste and grape marc is not augmented with alternative feedstock. With nearly 2.4 million tons of waste produced by dairy cattle annually [35] and only 8010 tons of grape marc produced annually, the treatment of all grape marc (at 35% of total treated biomass) would require appx. 1% of the dairy cattle manure provision capability of Oregon. However, the total production of this system might not be enough to provision a fully industrial scale biogas plant, though it would be enough to provision a smaller scale plant, and implications of this are discussed in Section 2.7.4.

Conversely, in relation to Oregon, the capacity of the biorefinery systems in Languedoc-Roussillon is limited by the production of manure. With only 18,700 dairy cattle [36], the region would only be able to supply appx. 0.37 million tons of the 0.39 million tons manure needed for co-digestion with the 0.21 million tons of grape marc produced in the region annually (CIVL—Conseil Interprofessionnel des vin AOC du Languedoc et des IGP Sud de France—Languedoc Wines). This relationship, unlike that in Oregon, is fairly well balanced. However, unlike in Oregon, there are well-established uses for grape marc, so the ability to provide grape marc as feedstock would therefore compete with existing demand (see Section 4).

Table 1. Feedstock provision for Languedoc-Roussillon and Oregon.

	Languedoc-Roussillon	Oregon
Annual Grape Marc Production (tons at crush)	212,940	8,009
Annual Cow Waste Production (tons)	372,300	2,389,091
Max. Co-digestion Feedstock Availability at 35% Grape Marc (tons/day)	1569	62
Cow Waste Demand at 100% Grape Marc Utilization (tons)	395,460	14,875
Grape Marc Demand at 100% Cow Waste Utilization (tons)	200,469	1,286,433
Cow Waste Demand at 100% Grape Marc Utilization (% of available cow waste)	106%	0.62%
Grape Marc Demand at 100% Cow Waste Utilization (% of available grape marc)	94%	16,061%

2.6. Impact Assessment Method

The ReCiPe 2016 Hierarchist method was used for impact assessment [37]. Impacts were assessed at the midpoint level with a time horizon of 100 years from the time of emission. All impact categories were included in the assessment of the dynamic system model and in all scenarios.

While all impact categories were modelled, using all indicators creates difficulty in relation to the interpretation of the results. To avoid this obstacle, GWP was chosen as a single indicator for impacts. In order to check for potential burden shifting when solely using GWP as an indicator impact, TOPSIS was applied with equal weighting to all impact categories. Ranking of the scenario results was then performed in a pairwise fashion, i.e., within each energy mix future, for the two scenarios, biogas-only and PHA-biogas, using both GWP as a single score indicator and TOPSIS.

2.7. Sensitivity Analysis

Important modelling parameters and assumptions were tested through a sensitivity analysis. These include:

2.7.1. Process Energy Consumption Related to PHA Production

Energy consumption related to PHB production was calculated using process design software, and it was subsequently tested to see if the overall results were sensitive to this parameter. Thus, a scenario where the energy consumption of PHB production does not improve over time was tested. For contrast, a scenario where processing improves by 5% per year was also explored.

2.7.2. Replacement Ratio Conventional Polymers

Replacement ratios of PHB to PET and PLA were estimated using the following material property indices: tensile strength, yield strength (σ), and the average between tensile strength and yield strength. RRs in the first model run were based on yield strength (σ), which applies to brittle polymers that are loaded in tension. This is done in order to relate the polymer matrix to its final application, which is unknown and is most likely several different applications for this case study. Thus, by choosing a handful of material properties, it is possible to estimate more realistic RRs that apply to desired properties. The values used of the RR estimation are presented in Table 2.

Table 2. Material properties, performance indices of polyethylene terephthalate (PET), polylactide (PLA) and polyhydroxybutyrate (PHB). Replacement ratios are derived from material properties using Equation (1).

	PET [38]	***PLA*** [39]	***PHB*** [40]
Yield strength, σ (Mpa)	2410.0	3830.0	2200.0
Tensile strength (Mpa)	38.8	48.0	32.0
Density (kg/m^3)	1.3	1.2	1.2
Performance index (YS)	1882.8	3088.7	1833.3
Performance index (TS)	30.3	38.7	26.7
Average performance	956.6	1563.7	930.0
Replacement Ratio (RR), YS	0.97	0.59	
RR, TS	0.88	0.69	
RR, AVG	0.93	0.64	

2.7.3. Mineralization of N in Digestate

An important source of uncertainty comes from the application of digestate to the field. In the first model run, EFs for N_2O emissions were based on IPCC values. To test the possible range of impact arising from N_2O emissions in the field, a powerful greenhouse gas, a second model run was performed using the N_2O emission factors published by [22]. Though these are not local EFs, they are used to portray the potential variation of greenhouse gas emissions after digestate application. The values used are found in Supplementary Table S5.

2.7.4. Feedstock Provisioning Scenarios

In both regions, there is potential for increased ground transportation induced by transport of grape marc for PHB production. Transport for grape marc is, in most cases, non-existent in Oregon whereas transport is used to distribute grape marc amongst various end-users in France. This means that implementing a PHA-producing biorefinery would either route or re-route the grape marc needed as feedstock to the biorefinery. To account for this, the system was modelled with ground transport of the grape marc by lorry. This was done for various potential transport distances ranging from 50–500 km for the PET replacement scenario.

3. Results

Results showed that the PHA scenarios outperformed the biogas-only scenarios in almost every impact category with a few exceptions (Figure 4). Exceptions included the French energy scenarios for the Ionizing Radiation (IR) impact category and almost all scenarios for Land Use (LU), except in one instance, the Oregon Static scenario, where PHA-biogas performed better than biogas-only in terms of LU.

It is worth noting that in some of the impact categories the difference between the two scenarios is so small that, keeping in mind the considerable uncertainty of LCA results in general, it is fair to say that both PHA-biogas and biogas-only are essentially equal in terms of environmental impact. This is true for the Particulate Matter (PM), Fresh Water Ecotoxicity (FWE), Land Use (LU), Marine Ecotoxicity (MEtox), Marine Eutrophication (ME), Mineral Resource Scarcity (MRC), both Ozone Formation categories, Terrestrial Acidification (TA), and Stratospheric Ozone Depletion (SOD) impact categories. The remaining impact categories show a greater degree of difference, where it is clear that the PHA scenarios are generally preferable. Midpoint impact category results are presented as percent reduction in environmental impact from the implementation of PHA production in relation to biogas-only scenarios, for all energy provision scenarios. These are shown both for scenarios replacing PET with a ca. 93% RR and a 30% RR, to show the influence of RR in impact results (Figure 5).

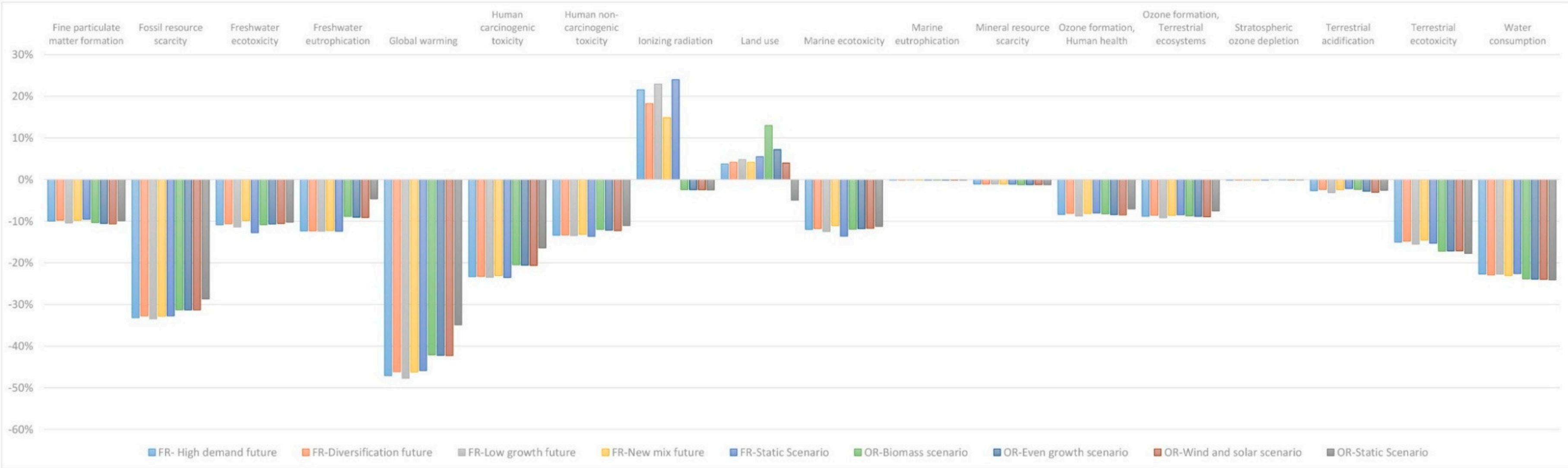

Figure 4. Relative difference for the PHA-biogas scenario. Negative values indicate that PHA-biogas outperforms Biogas-only scenarios.

	Fine particulate matter formation	Fossil resource scarcity	Freshwater ecotoxicity	Freshwater eutrophication	Global warming	Human carcinogenic toxicity	Human non-carcinogenic toxicity	Ionizing radiation	Land use	Marine ecotoxicity	Marine eutrophication	Mineral resource scarcity	Ozone formation, Human health	Ozone formation, Terrestrial ecosystems	Stratospheric ozone depletion	Terrestrial acidification	Terrestrial ecotoxicity	Water consumption
FR- High demand future	10.09%	36.88%	11.10%	14.86%	64.33%	27.53%	14.08%	-30.54%	-5.62%	12.23%	-0.13%	1.08%	8.62%	9.09%	-0.15%	2.70%	15.19%	22.78%
FR-Diversification future	9.78%	36.02%	10.79%	14.82%	61.39%	27.41%	14.00%	-28.86%	-6.19%	11.94%	-0.13%	1.09%	8.31%	8.78%	-0.14%	2.39%	14.89%	23.08%
FR-Low growth future	10.63%	37.46%	11.82%	14.94%	66.58%	27.80%	14.22%	-31.14%	-6.99%	12.89%	-0.13%	1.08%	9.08%	9.55%	-0.16%	3.24%	15.90%	22.80%
FR-New mix future	9.80%	36.13%	9.83%	14.68%	61.66%	27.04%	13.77%	-26.68%	-6.18%	11.10%	-0.13%	1.08%	8.36%	8.83%	-0.14%	2.42%	14.53%	23.32%
FR-Static Scenario	9.55%	35.99%	13.64%	14.96%	60.60%	27.91%	14.45%	-31.55%	-7.81%	14.40%	-0.13%	1.11%	8.17%	8.64%	-0.14%	2.17%	15.55%	22.60%
OR-Biomass scenario	10.52%	33.21%	11.07%	9.78%	50.11%	22.54%	12.19%	13.64%	-14.91%	12.13%	-0.15%	1.23%	8.47%	8.92%	-0.03%	2.39%	18.26%	24.50%
OR-Even growth scenario	10.77%	33.25%	10.89%	10.01%	50.44%	22.68%	12.46%	13.86%	-9.70%	11.99%	-0.15%	1.22%	8.68%	9.14%	-0.07%	2.86%	18.18%	24.64%
OR-Wind and solar scenario	10.92%	33.27%	10.73%	10.13%	50.63%	22.77%	12.61%	13.99%	-5.91%	11.86%	-0.15%	1.22%	8.79%	9.25%	-0.10%	3.12%	18.14%	24.72%
OR-Static Scenario	10.01%	28.67%	10.35%	4.65%	34.92%	16.41%	11.06%	14.30%	10.55%	11.29%	-0.15%	1.24%	7.08%	7.55%	-0.10%	2.60%	19.09%	24.93%

(A)

	Fine particulate matter formation	Fossil resource scarcity	Freshwater ecotoxicity	Freshwater eutrophication	Global warming	Human carcinogenic toxicity	Human non-carcinogenic toxicity	Ionizing radiation	Land use	Marine ecotoxicity	Marine eutrophication	Mineral resource scarcity	Ozone formation, Human health	Ozone formation, Terrestrial ecosystems	Stratospheric ozone depletion	Terrestrial acidification	Terrestrial ecotoxicity	Water consumption
FR- High demand future	-0.26%	13.73%	-0.31%	4.36%	29.86%	7.80%	3.55%	-36.84%	-14.44%	0.54%	-0.12%	0.14%	1.36%	1.53%	0.03%	-6.54%	2.63%	4.72%
FR-Diversification future	-0.55%	12.89%	-0.59%	4.33%	26.00%	7.69%	3.47%	-35.83%	-14.93%	0.29%	-0.12%	0.15%	1.07%	1.23%	0.04%	-6.82%	2.35%	4.98%
FR-Low growth future	0.23%	14.30%	0.34%	4.44%	32.95%	8.05%	3.67%	-37.19%	-15.59%	1.14%	-0.12%	0.14%	1.79%	1.95%	0.02%	-6.07%	3.27%	4.73%
FR-New mix future	-0.52%	13.00%	-1.45%	4.20%	26.35%	7.34%	3.26%	-34.52%	-14.91%	-0.47%	-0.12%	0.14%	1.12%	1.28%	0.04%	-6.79%	2.02%	5.20%
FR-Static Scenario	-0.75%	12.86%	1.99%	4.46%	24.99%	8.16%	3.89%	-37.42%	-16.27%	2.51%	-0.12%	0.16%	0.94%	1.10%	0.04%	-7.01%	2.95%	4.55%
OR-Biomass scenario	0.13%	10.18%	-0.34%	-0.30%	12.79%	3.24%	1.81%	-5.76%	-22.08%	0.45%	-0.14%	0.29%	1.22%	1.37%	0.15%	-6.82%	5.43%	6.28%
OR-Even growth scenario	0.36%	10.22%	-0.50%	-0.10%	13.13%	3.37%	2.05%	-5.58%	-17.82%	0.33%	-0.14%	0.28%	1.41%	1.56%	0.11%	-6.40%	5.36%	6.41%
OR-Wind and solar scenario	0.49%	10.24%	-0.65%	0.01%	13.34%	3.45%	2.19%	-5.47%	-14.66%	0.21%	-0.14%	0.28%	1.52%	1.67%	0.08%	-6.17%	5.33%	6.49%
OR-Static Scenario	-0.33%	5.96%	-0.99%	-4.91%	-1.62%	-2.10%	0.77%	-5.22%	-0.34%	-0.30%	-0.14%	0.30%	-0.07%	0.09%	0.08%	-6.63%	6.20%	6.68%

(B)

Figure 5. Percent reduction in environmental impact for all midpoint impact categories for the implementation of PHA production relative to biogas-only, for all energy provision scenarios with a PET RR of (**A**) 93% and (**B**) 30%.

The first model run shown in Figure 4 has PET as the conventional polymer to be replaced by PHB. The model was checked to see if a different polymer substitution material would alter the results. It was found that a change to PLA as the polymer substitution material did not change the general ranking, but the magnitude of the difference between PHA-biogas and biogas-only, i.e., the advantage that PHA-biogas has over biogas-only, decreased. Figures and tables for the PHA-biogas results for PLA are shown in the SI (Supplementary Figure S4 and Table S7).

Figure 6 shows the difference between the PHA-biogas and biogas-only scenarios, i.e., PHA-biogas CO_2-eq *minus* biogas-only, in CO_2-eq. For all 20 years, the PHA-biogas scenario induces greater savings than the biogas-only scenarios, which is why the results are always negative. Furthermore, the general negative slope of all scenario lines shows that as time progresses PHA-biogas becomes more attractive, inducing higher savings in comparison to biogas-only. More interestingly, it is possible to observe the difference between plans for energy grid development in the two locations. Hence, Oregon scenarios show a steeper slope, i.e., a drastic pull back from the use of fossil fuels and, more specifically, the use of coal. In contrast, the French slopes are less pronounced, as improvements to the grid are subtler because there is already a large share of non-fossil-based energy production in use in France. The difference between the two regions is larger at the beginning of the period, getting smaller in time as the grids progressively increase their share of renewable energy.

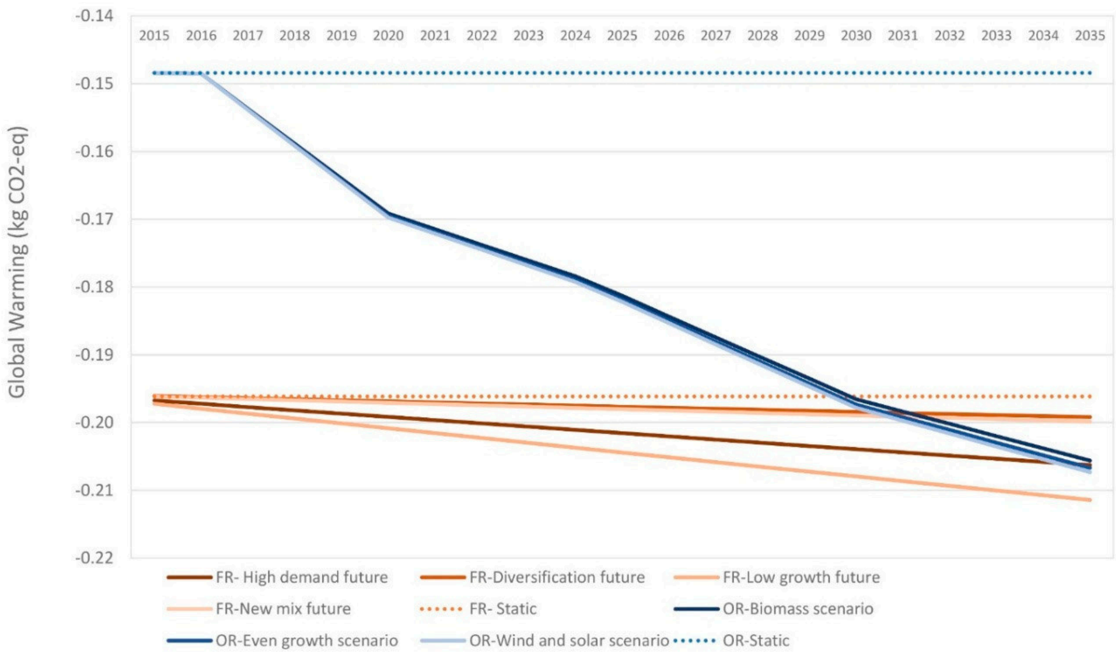

Figure 6. Yearly difference of global warming potential (GWP) impacts, i.e., PHA-biogas minus biogas-only scenarios. Figure reflects the evolution of the energy mixes in the two locations. Negative values mean PHA-biogas has higher savings than biogas-only.

3.1. Sensitivity Results

The robustness of model results was checked by varying different parameters, as described in the methodology, Section 2.7. After each change, indicators were checked with the TOPSIS and GWP single indicators, but for the most part, there was no change to the preference ranking of the scenarios, and combined PHA-biogas production continued to perform better. Thus, it can be said that the model results are robust in regards to the most influential parameters analyzed.

In more detail, changes to the replacement ratio (RR), i.e., the PHB: PET mass ratio that is allowed by different material properties, as discussed in Section 2.7.2, was shown to be a moderately sensitive parameter. A 5% change in the replacement ratio lead to a 3–4% change in results for PHA-biogas with PET (Figure 7), and a 2.5–4% change in results for PHA-biogas with PLA. Thus, it can be said that a general trend is observed of lower savings with lower RR (or higher savings with higher RR), while the effect of the change is nearly proportional to the change seen in the results.

The sensitivity to efficiency improvements for PHA-producing technology was also tested and it is shown in the SI, Figure S3. This parameter was showed to have very little effect on overall model results, with GWP changing in the range of 0.1–1.5%.

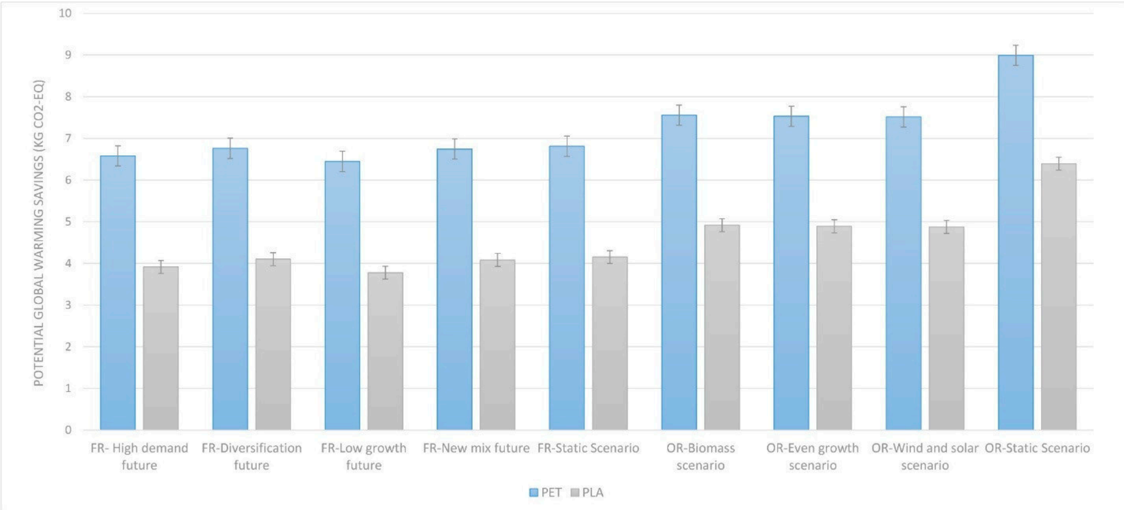

Figure 7. Sensitivity analysis of replacement ratio of conventional polymers by PHB. Cumulative GWP savings for substitution of PET, in blue, or PLA, in gray, by PHB. Bars represent savings in relation to the biogas-only scenario and the upper and lower error bars represent the range of potential savings, which depends on material properties' performance indices. PHA scenarios only.

Sensitivity of N_2O Emission Factor

Cumulative global warming impacts switch from a savings inducing status to a burden inducing status when N_2O emission factors for the field application of digestate from [22] are applied (Supplementary Figure S4). However, the ranking between PHA-biogas and biogas-only stays the same, with combined PHA-biogas scenarios continuing to perform better than biogas-only scenarios. The results show that N_2O emissions play an important role, and considering the strong dependency on local conditions, they should as much as possible be spatially differentiated. The variability of N_2O emissions for the EFs employed can be seen in Figure 8.

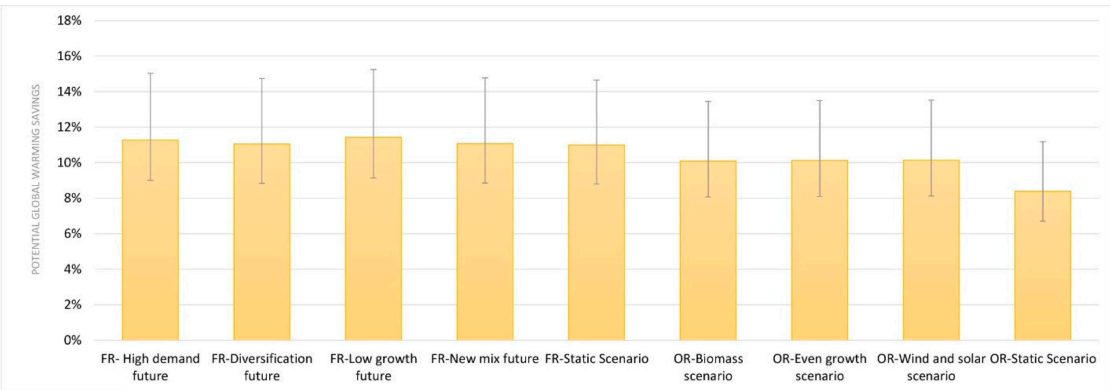

Figure 8. Cumulative PHA-biogas GWP minus cumulative biogas-only GWP. Yellow bars indicate relative savings of PHA-biogas scenarios in relation to biogas-only for each energy mix future. Error bars indicate variation in the savings induced by PHA-biogas due to N_2O emissions after application of digestate. Upper error bars correspond to the high crop response case, while lower error bars correspond to the low crop response case, as explained in Section 2.3.2.

3.2. Territorial Scale Application

Application of the biorefinery alternatives at a territorial scale would lead to potential reductions in regional environmental impact. In order to give a measure of scale to the potential savings induced by the implementation of maximum (limited by feedstock availability) PHA-biogas production relative to biogas-only, the GWP impacts were normalized using planetary boundary carrying capacity-based normalization factors [41]. Assuming a 985 kg CO_2 eq. per person year (PY) carrying capacity (C.Cap) [41], and assuming that PHA replaces PET with a 93% RR and that the PHA process improves in terms of energy efficiency at 1% annually, the production of PHA induces an average reduction in GWP impacts relative to biogas-only equating to nearly 1400 PY of C.Cap. When broken down by region, the French scenarios indicate an average relative maximum potential GWP saving of over 2400 PY of C.Cap, with Oregon exhibiting just over 80 PY of C.Cap in average relative maximum potential GWP savings. Using the same assumptions, except exchanging the replacement polymer with PLA production at a 64% RR, then the maximum implementation in France and Oregon of the PHA-biogas scenario induces an average annual potential relative GWP impact reduction of 493 PY of C.Cap when compared to production of biogas-only, with 871 and 21 PY of C.Cap in France and Oregon, respectively, see Table 3.

Table 3. Carrying capacity normalized GWP reduction for maximum application of the PHA-biogas relative to the biogas-only biorefinery alternative in France and Oregon based on replacement of PET with 93% RR and a 1% annual energy efficiency improvement for PHA production. Reduction per functional unit (FU).

	GWP (Kg CO2e) Reduction/Fu	Person Years (PY) of Carrying Capacity (C.Cap) Reduction Daily	PY of C.Cap Reduction Annually
FR-HIGH DEMAND FUTURE	4.23	6.74	2460.75
FR-DIVERSIFICATION FUTURE	4.15	6.61	2413.15
FR-LOW GROWTH FUTURE	4.29	6.84	2495.46
FR-NEW MIX FUTURE	4.16	6.62	2417.67
FR-STATIC SCENARIO	4.13	6.57	2399.86
OR-BIOMASS SCENARIO	3.79	0.24	86.98
OR-EVEN GROWTH SCENARIO	3.80	0.24	87.25
OR-WIND AND SOLAR SCENARIO	3.80	0.24	87.41
OR-STATIC SCENARIO	3.14	0.20	72.14

Sensitivity Analysis of Transport at Territorial Scale

The importance of transport was tested via sensitivity analysis of different theoretical grape marc transport distances for both the biogas-only and PHA-biogas scenarios (Table 4). For all scenarios, a 500 km transport distance results in overall elimination of environmental benefits, and at 200 km, transport of grape marc reduces average impact savings from the various biorefinery-region scenarios by 42.5% for all midpoint indicators. In terms of GWP, a 200 km transport distance induces impacts of a maximum of appx. 284% and a minimum of 68% of the magnitude of GWP savings without transport. At 50 km, all scenarios show reductions in GWP. At 100 km, all PHA production scenarios and France biogas-only scenarios induce GWP savings, while the Oregon biogas-only production scenarios eliminate the GWP benefit of implementing the biorefinery. Furthermore, if the introduction of centralized PHA-Biogas biorefineries were to induce transport of grape marc, relative to existing decentralized biogas production, then GWP savings are overwhelmed by the induced impact from transportation at any distance greater than appx. 125 km.

Table 4. Sensitivity to inclusion of transport of grape marc in percentage change to midpoint impacts without transport.

	50 km	100 km	200 km	500 km
AVERAGE CHANGE AMONGST ALL IMPACT CATEGORIES	11%	21%	43%	106%
AVERAGE CHANGE IN GWP	36%	73%	145%	363%
MAX. CHANGE IN GWP	71%	142%	284%	710%

4. Discussion

Overall, the model results obtained were robust and indicate that implementing PHA production technology is preferable to conventional anaerobic digestion, when the functional unit (FU) equals 1 ton of feedstock treated. Combined PHA-biogas scenarios, whether with PET or PLA as the replaced polymer, performed better across almost every impact category. This is largely due to the added benefit of replacing conventional polymers, which are associated with significant impacts. As evidenced by the replacement ratio (RR) sensitivity analysis, decreasing or increasing the amount of PHB needed to equate the function of PET or PLA resulted in an almost proportional effect in the outcome. RR of PET would have to decrease by around 80% and be as low as 20% before there is rank reversal between the two options in some of the impact categories. This was confirmed by both single score indicators, which prefer combined PHA-biogas scenarios until reaching values close to 20% RR (Table 5). However, the GWP single indicator still preferred PHA-biogas, even at a 20% RR, except for the OR-Static Scenario. On the contrary, the TOPSIS single indicator, which is equally weighted between impact categories, starts preferring biogas-only scenarios earlier, with a 35% RR. In this regard, there was less operating space for the GWP indicator, when PLA is the replacement polymer, which starts signaling biogas-only as the preferred choice already at 30% RR. On the contrary, TOPSIS selects biogas-only at low RR of 9–16%. Thus, there is disagreement between the GWP and TOPSIS single indicators, which is, furthermore, replacement polymer-dependent. This points to two issues to consider: (1) choosing GWP as the only impact category for the assessment can potentially result in burden shifting to other environmental impact categories and (2) the choice of polymer substitution affects impact categories other than GWP, here exemplified by the difference in the TOPSIS results when choosing PET or PLA as polymer replacement. To elaborate, the difference lies in PET's production being more burdensome for impact categories other than GWP in comparison to PLA's production. However, the single score indicators employed generally indicated a similar scenario prioritization, i.e., combined PHA-biogas production being the preferred choice across all future energy scenarios, as long as RRs were higher than 20% for PET and 30% for PLA. It is worth noting that such a low replacement ratio is considered unrealistic, as the material properties of PHB allow for various applications [40].

Table 5. Single indicator preference, by TOPSIS with equal weights or GWP. Sensitivity values shown. For energy demand of calculated PHA production, values start with 10 times the calculated energy needed. For RR, values are shown for a replacement rate lower than 42%; above this value, PHA-biogas is always preferred.

		FR-High Demand Future	**FR-Diversification Future**	**FR-Low Growth Future**	**FR-New Mix Future**	**FR-Static Scenario**	**OR-Biomass Scenario**	**OR-Even Growth Scenario**	**OR-Wind and Solar Scenario**	**OR-Static Scenario**
Energy Demand for PHA Production (kWh/FU)										
70.70	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
77.70	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	PHA	PHA	PHA	PHA	Biogas	PHA	PHA	PHA	PHA
84.84	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	Biogas	Biogas	Biogas	PHA	Biogas	Biogas	PHA	PHA	PHA
98.98	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	PHA	PHA	PHA
106.10	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	PHA	PHA
113.12	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	PHA	PHA
127.26	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	PHA
226.34	GWP Preference	PHA	PHA	PHA	PHA	PHA	Biogas	Biogas	Biogas	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas
388.85	GWP Preference	PHA	PHA	PHA	PHA	Biogas	Biogas	Biogas	Biogas	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas
537.32	GWP Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas
Polymer replacement ratio (PHB:PET)										
42%	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
32%	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	PHA	PHA	PHA	PHA	Biogas	Biogas	Biogas	Biogas	PHA
22%	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas
12%	GWP Preference	PHA	Biogas	PHA	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas

Much like with polymer replacement ratios, TOPSIS and GWP do not always agree when the limits of process energy consumption are tested. If process energy consumption reaches 134 kWh per FU of added energy demand for PHA production, then TOPSIS (unlike GWP) indicates preference for biogas-only, for all energy scenarios, which indicates there is a potential for burden shifting if GWP is chosen as the only indicator. However, unlike the replacement ratio, improvements in process energy consumption for the production of PHA lead to very small changes in results. If there is no improvement in process energy consumption, meaning production of PHA consumes 7 kWh more per FU than the biogas-only scenario, results still stay the same. The break-even point of energy consumption for PHA production is high, i.e., it takes 12 times this value, 85 kWh of added process energy consumption of PHA per ton feedstock, before the TOPSIS-derived single indicator shows preference for biogas-only over combined PHA-biogas production for several of the French energy scenarios and one Oregon scenario. Moreover, it takes 16 times this value, or 113 kWh/FU more, before it is possible to observe prioritization change for the GWP single indicator for one Oregon scenario, the OR-Static Scenario, and 32 times the initial value, 226kWh/FU, before all Oregon energy scenarios show a preference for biogas-only. As for France, it is not until PHA production consumes 55 times this value, 389 kWh/FU, before there is a change in the GWP single indicator in preference of one of the energy future scenarios; the FR-Static Scenario. Thus, it is possible to conclude that there is large leeway in process energy consumption for PHA production before the decision support will change, in terms of GWP. As exemplified here, this is also dependent on the share of renewable energy sources in the future energy grid, which is why results are more robust for France in terms of GWP, i.e., requiring 55 times, 7 kWh/FU, more energy consumption before seeing a change in GWP impact category. The energy prediction mix is thereby an important factor when deriving the impacts of the system, which are heavily affected by energy mix usage.

In this regard, using dynamic energy grids for the background is a powerful tool. Many nuances are highlighted and originate from the predicted/expected changes in the share of renewable energy for the different locations. The most obvious of these subtleties can be observed in the Ionizing Radiation category (Figure 4), where it is evident that there is a higher share of nuclear energy in the French background system than in that of Oregon. As seen in Figure 6, the evolution of the energy grid reveals a sharp decrease for Oregon, while France's energy grid remains somewhat unaltered. This is due to legal requirements in Oregon, which are intended to increase the share of renewables from 15% to 50% by 2040 [28]. Greening of the energy grids increases the difference between biogas-only and PHA-biogas in the future, as is exhibited by the negative slopes of the lines in Figure 6. Despite the increasing environmental importance of plastic replacement as opposed to electricity replacement, it is worth restating that PHA-biogas is consistently preferable in terms of GWP, i.e., negative values throughout the assessment period. One major area discussion regarding the dynamic inventory is the use of local energy mix scenarios in commodity replacement. It is likely that the increased production of PHA would have no direct effect on the production of PET or PLA in Oregon or France. However, by using a local instead of global process, it is possible to develop processes that are treated equally, in terms of system dynamism, for their inventory development. Furthermore, this is seen as a cautious choice, as the localized dynamic processes for the replaced polymers exhibit lower impacts than the global average. Thus, it is possible that this inclusion slightly under-represents the potential impact reduction gains from increased PHA production and is hence considered unlikely to over-state impact reduction gains.

As shown in the sensitivity analyses, biogas-only scenarios are preferred only in extreme cases where polymer replacement ratio or consumption of energy during PHA production are set to extreme values, i.e., very low RR and very high process energy consumption for PHA. Another area of uncertainty is N_2O emissions after digestate application, which have also been shown to be highly uncertain in several LCAs [42–44]. N_2O emissions were shown to have the potential to induce impacts for all scenarios, though the ranking of PHA-biogas in relation to biogas-only was not affected. Due to the closeness in results from the field application of digestates generated from the model for biogas and PHA scenarios, it can be concluded that both digestates act more or less in the same way during field application. Results were also tested without the field emissions, leading to the same technology prioritization. Nevertheless, it is important to highlight the large impact that N_2O emissions have in assessing agricultural product systems, and the necessity to improve inventories of these emissions in LCA assessments. Incidentally, the TM-LCA framework advocates for the use of local inventory data as much as possible.

One area that is made evident by including the territorial assessment, where there is potential for inducing impacts that would eliminate the environmental benefits of the system, is transport. Due to the relatively low energy and chemical value density in grape marc, increases in present transport of grape marc greater than 200 km cause induced impacts in all biogas-only scenarios. When transporting grape marc 250 km, both PHA-biogas with PET replacement and biogas-only induce impacts, except for the PHA-biogas scenario with static energy grid in Oregon, i.e., a dirtier energy mix than impacts from transport. Furthermore, if the PHA-biogas scenario induces transport relative to the biogas-only scenario (no added transport for biogas-only), then 150 km of grape marc transport eliminates the GWP benefit of the PHA-biogas scenario. While the PHA production scenario remains clearly preferable to biogas-only in all transport scenarios, this result does underline the need to assess potential re-routing of the feedstock if a new biorefinery technology were to be implemented.

It is also notable that the present use of feedstock, omitted in the results of this study as the impacts would be equal in both the PHA-biogas and the biogas-only scenarios, varies significantly between the two assessed territories. In France, there is a well-established market for distillation of wine residues, and in Oregon the wine residues are often used as compost. This said, it is also important to highlight that the feedstock mix used in this assessment can also be changed, as the PHA-producing technology is compatible with all types of organic waste, e.g., the organic fraction of household waste, waste-water treatment sludge, other animal slurries, other crop residues etc. The option to change the feedstock mix was not investigated in this study, as it would change the functional unit and was thus omitted from the present work. However, it is quite possible that there is further exploitable feedstock in both assessed regions. A good indication of feasibility is if there is an industrial sized biogas plant already in operation in the region; this would indicate that there is already feedstock enough to run PHA production. However, it is important to keep in mind that the use of crops has not been investigated in this report and so this study's conclusions do not apply if the feedstock is food crops.

5. Conclusions

Based on the results of this study, it can be concluded that when a biorefinery is installed in Oregon or Languedoc-Roussillon to handle a mix of grape marc and cow waste, it is very likely that it would be environmentally beneficial to include PHA production in addition to energy and digestate production. When relating the impact reductions between PHA-biogas and biogas-only, based on the maximum potential implementation capacity of the specific region, to planetary boundaries-based carrying capacity, it is shown that the impact reductions correspond to up to nearly 2500 person years in France and up to nearly 90 person years in Oregon. This corresponds to 1.59 and 1.40 person years of avoided GWP per ton of treated feedstock per day in France and Oregon, respectively. However, based on the results of the sensitivity analysis regarding transportation, special care needs to be taken in regards to assessing the potential increase in biomass transport; otherwise, it is likely that all environmental benefit from the biorefinery will be offset by the induced impacts of transportation. Likewise, the induced environmental impact reductions cannot be ensured if the feedstock for the biorefinery is to be rerouted from another use. Thus, it is concluded that PHA production should be seen as a potentially valuable add-on for biogas platforms.

The TM-LCA framework has the added benefit of elucidating the influence of potential future energy provision and the impact this has on potential environmental benefits. As indicated by the results, the benefit of including co-production of PHA in biogas plants increases as energy grids become greener, an element that can have significance in terms of decision support for its implementation from the regional planning or governance perspective. The framework also provides perspective on the scale of potential benefits (in person years) and added emphasis on single score indicators that point out possible burden shifting to environmental problems other than global warming.

Supplementary Materials: The following are available online at http://www.mdpi.com/2071-1050/11/14/3836/s1.

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Data Availability: Data related to this publication is available by clicking on the URL, through the INRA dataverse, which is available as OpenAir+ EU datawarehouse. (https://data.inra.fr/privateurl.xhtml?token=42512e88-61e9-40ba-965c-578a611eefb0).

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Assessing New Biotechnologies by Combining TEA and TM-LCA for an Efficient Use of Biomass Resources

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Abstract: An efficient use of biomass resources is a key element of the bioeconomy. Ideally, options leading to the highest environmental and economic gains can be singled out for any given region. In this study, to achieve this goal of singling out an ideal technology for a given region, biotechnologies are assessed by a combination of techno-economic assessment (TEA) and territorial metabolism life cycle assessment (TM-LCA). Three technology variations for anaerobic digestion (AD) were assessed at two different scales (200 kW and 1 MW) and for two different regions. First, sustainable feedstock availability for two European regions was quantified. Then, the environmental impact and economic potential of each technology when scaled up to the regional level, considering all of the region's unique sustainably available feedstock, was investigated. Multiple criteria decision analysis and internalized damage monetization were used to generate single scores for the assessments. Preference for the technology scenario producing the most energy was shown for all regions and scales, while producing bioplastic was less preferable since the value of the produced bioplastic plastic was not great enough to offset the resultant reduction in energy production. Assessing alternatives in a regional context provided valuable information about the influence of different types of feedstock on environmental performance.

Keywords: anaerobic digestion; polyhydroxyalkanoates; life cycle assessment; techno-economic assessment; territorial metabolism; regional assessment; wet oxidation; biogas; biomass valorization

1. Introduction

One of the goals of the European Union (EU) is to stimulate the creation of a competitive low carbon economy that is able to provide a reduction of 80%–95% greenhouse gas (GHG) in Europe by 2050 [1]. Energy production is an important sector where changes can be made in order to reach this ambitious target. Shifting from fossil-based energy to renewable sources of energy can lead to GHG reductions, provided the value chains for the renewable energy sources can lead to better overall environmental performance. A careful evaluation of new renewable energy pathways has previously been recommended [2] and various studies have shown wide ranges for GHG emissions of renewable energy systems [3,4]. Moreover, and particularly relevant to biomass based renewable energy, in some cases lower GHGs are not accompanied by lower emissions of other environmentally concerning emissions, such as those contributing to eutrophication, acidification, and human/ecosystem toxicity [5,6]. Within the various renewable energy sources, biomass is important as in 2015 it already supplied 10% of the global demand for primary energy consumption [7]. In Europe, demand for electricity biomass, heating, and transport was around 5010 PJ in 2012 and it is estimated to rise to 7437 PJ in 2020 in order to meet renewable energy targets in the EU. Thereby, it is important to consider additional renewable energy with holistic perspectives that can quantify the environmental performance of renewable energy from biomass resources. Life cycle assessment (LCA) is an internationally recognized, standardized tool with a mature methodology capable of assessing large systems and giving a complete assessment of environmental impacts [8]. As such, LCA has been used widely and is aligned with the sustainable development goals (SDGs) developed by the United Nations [9], which incorporate life cycle thinking into, for example, goal number 12 (sustainable production and consumption patterns) [10]. Under the umbrella of SDGs, decoupling economic growth from the unsustainable use of resources is of prime importance so that future generations may enjoy precious natural resources. Thus, measuring progress towards these goals is necessary from both an economic and environmental perspective, which makes the use of mixed assessments necessary.

Out of the estimated 7437 PJ demand for biomass energy in 2020, 887 PJ are expected to come from biogas [11]. Biogas production has increased significantly in the EU, from 92 PJ in 2000 to 654 PJ of primary energy in 2015, with a total of 17,400 installed biogas plants [7]. Anaerobic digestion (AD) is a versatile technology for many reasons, one being that it is possible to install decentralized plants near agricultural sources of feedstock. In terms of biomass resources, AD can utilize various types of organic waste aside from agricultural residues, including industrial wastes such as slaughterhouse wastes and residues from food production, sewage sludge and the organic fraction of municipal solid waste. The produced biogas can be valorized in several ways, such as for heat and electricity production in combined heat and power engines (CHP); injection into the natural gas grid after an upgrade to biomethane; or use in the transport sector. It is at least in part due to this versatility that AD can serve as a successful platform for the bioeconomy. In addition, the latest developments in biogas technology expand the platform beyond energy into materials production [12]. While some of the advances focus on optimizing energy extraction, such as wet explosion pretreatment aimed at unlocking the lignocellulosic fraction of waste [13], or adding a separate dark fermentation step before methanization so as to increase hydrogen content of the biogas [14], other innovation allows for the production of biopolymers via the modification of the AD process [15]. By isolating the volatile fatty acids (VFAs) produced during the AD process and feeding them to microbes in a multi-stage process, intracellular polymer, such as polyhydroxybutyrate (PHB) of the polyhydroxyalkanoates (PHAs) family of biodegradable polymers can be produced and later extracted from the bacteria. In this way, it is possible to turn biogas plants into chemical platforms, which can expand the acting field of AD to new utilization and valorization opportunities.

Needless to say, biogas relies on available biomass and by definition is constrained to these finite resources. Various studies have focused on mapping out the availability of biomass in Europe for the production of energy and biogas [16–21]. Though the quantified potentials vary widely due to methodological selections and database choice, it is generally acknowledged that the extraction of biomass must be done with care to avoid competition with food resources and unwanted market effects, such as increases on land and maize prices [22,23]. Still, Scarlat et al. [11] warns that even though domestic biomass supply in the EU is enough to satisfy the demand required to accomplish national renewable targets, as much as a quarter of the biomass demand may be sourced from third countries (outside of the EU) in 2020. Since this is due to market effects, it is imperative to take economics as well as environmental aspects into account so that the appropriate support systems are in place for the development of a sustainable renewable energies market and thereby a sustainable biogas sector. In this regard, it is important to determine if the emerging biogas innovations mentioned are environmentally sound and lead to environmental performance improvements in comparison to the status quo. As has been pointed out before, the prefix bio does not guarantee sustainability [24]. Biogas capacity already built in Europe is an important aspect when analyzing any additional capacity that may be built in an area, e.g., considering that 50% of the EU's biogas capacity is in Germany [7]. As has been pointed out by Bojesen [25] and colleagues, who estimated service areas for existing and future biogas plants in Denmark, the availability of feedstock in relation to plant location is an important aspect. An inadequate assessment of a plant's sourcing ability may lead to high operation costs from increased transport demand or inadequate sourcing of feedstock [25]. In turn, high transport distances may negate the environmental benefits brought about by biorefineries, as shown in Croxatto Vega et al. [26] which applied the territorial metabolism-LCA approach (TM-LCA) [27] and found distances of 50 km to be the upper limit.

This study performs a step-wise assessment starting from individual plant level and investigates the implementation potential of the PHB and AD-Booster technologies in two different plant scales. A techno-economic assessment (TEA) and LCA are carried out for this aim. The results from the TEA-LCA are used to structure implementation of the technologies at the regional level. The TEA relates the plant scale and processing capacity to capital expenditure (CapEx) and operational expenditure (OpEx) of the plant, and to the break-even prices of products. In the LCA, the environmental aspects of different technologies are quantified. The implementation of the two technologies is analyzed for two regions defined by the nomenclature of territorial units for statistics (NUTS) from Eurostat's definition of regions (NUTS2 regions): Bavaria, Germany and Veneto, Italy. We analyze the potential impacts of the two innovative technologies (PHB and AD-Booster) against the current level of biogas implementation for the regions. First, we use TEA to analyze the effect of scale on the economic potential considering relevant plant sizes. Concurrently, we provide a mass flow analysis for the regions to better understand the energetic potential of agricultural residues produced within the regions (i.e., both the residues already in use for biogas and not yet exploited) as well as the level of development of the biogas sector (i.e., installed capacity). Finally, we use the results from the TEA of each technology to perform a TM-LCA, which will be able to tell us the possible environmental improvements (or deterioration) potentials for the whole region, if all of the residues are processed with the new technologies. We place special attention on the repercussions for the farmer, especially from installation of large biogas plants, which can potentially monopolize biomass resources over a large area. Vice versa, we explore the possible needs and constraints for biogas developers in the two regions. In this way, we seek to explore new biotechnological implementation potentials from a stakeholder's perspective.

2. Method

2.1. Plant Level Assessment

The potential of implementing new AD technology was analyzed at two different scales. Data was collected from two biogas plants: a 1 MW installed electric capacity plant in Veneto, Italy and a smaller 200 kW plant in Bavaria, Germany, hereafter referred to as "the farms". Both plants operate on a mixture of cow manure, crop residues, and maize silage (Table 1). Both plants valorize biogas in CHP units, which produce heat as a waste product. Both plants utilize the co-produced heat in the plant's operation and additionally, in the Bavarian case, the surplus heat produced is utilized in the district heating network for a nearby village [28].

Table 1. Feedstock mix employed in the farms.

	200 kW		**1 MW**	
	% ww [1]	**ton/day**	**% ww [1]**	**ton/day**
Cow manure	57%	11.3	82%	131.4
Maize Silage	27%	5.5	14%	23.0
Grass silage	14%	2.7	3%	5.4
Grain	2%	0.4	0%	0.0

[1] Percent on a wet weight basis.

2.2. Technology Description

Three technology scenarios were assessed. Conventional AD was chosen as the baseline and two emerging treatment processes that can be added to existing AD were assessed for the comparison. All technology scenarios are modelled with a biogas leak of 3% of the produced biogas [22]. The technology set ups are: AD, AD + Booster, and AD + PHB.

2.2.1. Anaerobic Digestion

Conventional AD was modelled using SuperPro Designer, following the details received from the farms (Figure 1). The feedstock is grinded before it enters the anaerobic digester. The anaerobic digester produces biogas and digestate. The AD model was populated with the most common stoichiometric equations governing anaerobic digestion in [29]. Internal electricity consumption for the whole process was 7.5% of produced electricity based on data obtained from the farms operating biogas plants. A methane content of 50% for the biogas and an electrical efficiency of 38% for the CHP unit was used, based on the received data, yielding a 1.9 kWh/m^3 of biogas. Internal thermal energy usage was assumed to be 40%. The methane content, electrical efficiency, energy content of the biogas, and internal heat use was equal in all technology scenarios.

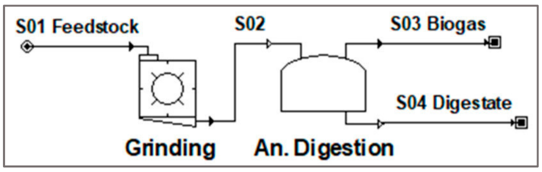

Figure 1. Simplified flow chart of anaerobic digestion.

2.2.2. AD + Booster

The AD + Booster technology consists of an extra tank where the wet explosion technology is applied under high heat and pressure conditions [13]. The AD + Booster scenario (Figure 2) was designed with information obtained from the technology developers [30]. In comparison to AD, the AD + Booster technology increases the conversion yield of cellulose to biogas from 52% to 88% and the conversion yield of hemicellulose to biomass from 75% to 98%. This scenario has an internal electricity consumption of 9.5% of produced electricity. On the other hand, the biogas yield is 12% to 16% higher than the AD scenario.

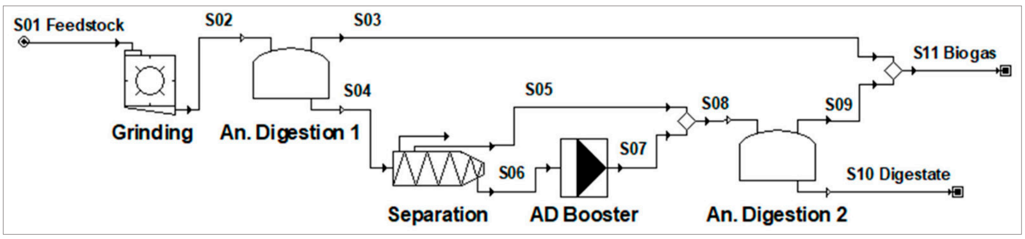

Figure 2. Simplified flow chart of the anaerobic digestion (AD) + Booster technology.

2.2.3. AD + PHB

In order to include a PHB producing section into an existing AD plant, a few extra pieces of equipment are necessary (Figure 3). AD is split into two tanks, the first is of short retention time and is where the VFA are produced and rerouted for PHB production. After this step, a screw press and a filtration unit separate solid from liquid. The solid fraction is fed to the AD tank where it continues the regular AD process, while the liquid fraction goes into a series of bio-oxidation units where selection and accumulation occurs via the feast and famine method [15]. The bio-oxidation equipment, in SuperPro Designer, was populated with stoichiometric equations obtained from the technology developers. Finally, PHB can be extracted using sodium hypochlorite and a final filtration step recovers a crude PHB. In comparison to AD, this scenario has an internal electricity consumption of 15% of produced electricity and a biogas yield from 24% to 30% lower than AD.

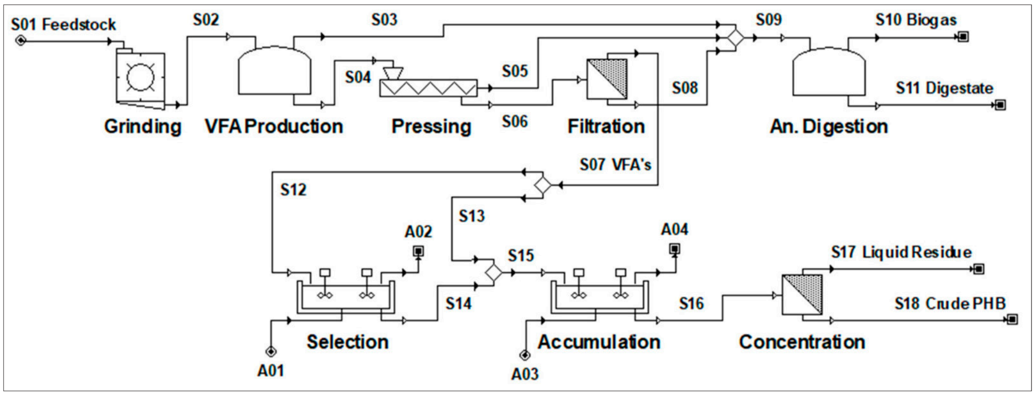

Figure 3. Simplified flow chart of the AD + polyhydroxybutyrate (PHB) technology.

2.3. *Regional Feedstock Availability*

2.3.1. Crops

Primary production amounts and land cover of individual crops was obtained from the Eurostat database (apro_cpnhr, [31]) for the two NUTS2 regions. As much as possible, the most recent data on production statistics was used, for the period of 2008–2018. For Veneto, data coverage on crops by Eurostat was very incomplete. Thus, it was preferable to use data from the National Italian Institute of Statistics (ISTAT) [32], where data is available for the whole period at the NUTS2 level. The production yield (production amounts divided by area of production) was then averaged over the period to derive an average production per year for the regions. Residue:crop ratios where then applied to the production yield to derive a total annual amount of residues for each crop. A list of the residue:crop ratios (Table 2), as well as grouping of Eurostat categories used are provided (Table A1, in Appendix A). For the most part, it was assumed that only residues are used for AD (or AD + innovation), with the exception of energy crops where the whole plant is assumed to enter digestion. Energy crops included are maize silage, green maize, and sorghum. Only crops which are most commonly considered for biogas operation were included in the study; we excluded horticultural crops that do not typically serve a purpose in AD.

Because wine grapes are an important crop in the Veneto region, they are also present, though to a smaller extent in the crop mix of Bavaria. Amounts of winery pomace were also taken into account as potential AD feedstock. Data on wine production was obtained from Eurostat (vit_t1, vit_an5, vit_an7, [33,34]) for both regions and the data for Veneto was checked against ISTAT data. The period for Eurostat wine data differed slightly and included the years 2001–2009, and 2015. The amount of pomace produced was estimated based on [35], which reports a 25% conversion rate from grapes to pomace.

After obtaining total crop residue amounts for the region, it is necessary to estimate a technical and sustainable potential for collection of the residues. The technical potential may potentially exclude the share of residues which is too difficult to collect, as well as the share that has known competing applications (Table 3). For example, this is the case for straw from cereal production, which is typically used for bedding and feed for cattle and other animals [36]. Sustainable potential collection, on the other hand, takes into consideration soil fertility. Residues may, for example, be left on agricultural fields to uphold the organic matter content of the soil and protect it from excessive erosion. Sustainable potential collection factors typically used in the literature vary from around 10% to 50% of the most common types of residues, i.e., excluding pomace and energy crop [37], and it has been shown that residue removals above 50% may negatively affect soil organic carbon storage [38]. Nearly all studies [11,16,19–21,36,39] that evaluate biomass potential for bioenergy purposes apply some sort of technical/sustainable collection factor, yet many of these studies do not report the actual values used or leave values out. We report all the values used in Table 2, since this is one of the most important determinants of potential for biomass utilization.

Table 2. Sustainable removal factor of various crops.

	Fraction of Total Residues	References
Cereal straw	0.3	[40]
Rice straw	0.5	[19,21]
Maize	0.5	[19,21]
Leguminous	0.1	[36,41]
Sugar neet	0.5	[36,41]
Rape	0.5	[19,21]
Sunflower	0.5	[19,21]
Soya	0.4	[17]
Oily	0.1	[36,41]
Industrial	0.4	[17]
Forage	0.1	[36,41]
Energy crop	0.9	[19,21]
Pomace	0.99	[42]

Table 3. Competing application factors for cereal straw.

	Feed [1]	Bedding [1]
Straw for bovine [2]	0.1	0.8
Straw for swine [2]		0.6
Straw for sheep [3]	0.025	0.2
Straw for goat [3]	0.025	0.2
Straw for poultry [4]	0.0125	0.1

[1] unit is ton per livestock unit*yr. [2] [40]. [3] Estimated value. Sheep and goat use a fourth of bovine. [4] Estimated value. Poultry uses a half of sheep and goat.

2.3.2. Manure

Animal production data was obtained from the Eurostat database (agr_r_animal) for bovine, swine, sheep, and goats for the period of 2008–2018. At the NUTS2 level, it is possible to obtain data for the number of animals in thousand heads. It is then necessary to estimate the amounts of manure excreted by the different types of animal, which varies also with their age (dairy cows, calves, sows, piglets, etc.). Values of manure production are calculated using the methodology detailed in [43] following the definitions for the various animals in [44]. The values are reported in Appendix A, in Table A2. Poultry production is not reported in the above-mentioned database, thus it was necessary to use the ef_lsk_main Eurostat database, which reports livestock units (LSU) for poultry for the years 2005, 2007, 2010, 2013, and 2016 at the appropriate regional level. This was the best available data for poultry at the NUTS2 level. LSU values were converted to poultry heads, following the methodology outlined in [43].

Similarly to crop residues, a technical potential was considered for animal manures. Here, for cattle, the potential collectable manure was estimated based on the type of housing and rearing. Since European regulation on organic production of agricultural products specifies that organic "livestock should have permanent access to open air areas" in most cases [45] and that there shall be a connection between land management by the use of manure, i.e., meaning that organic production must maintain the fertility of soil by applying cover crops, green manures or organic livestock manure, it was assumed that manure could only be collected in the harsh winter months (at most) from organic cattle farms [46]. The estimate for housing types was derived from the Farm Structure Survey (FSS) [47] carried out in 2010, since more recent FSS could not be located. The types of housing were assumed to stay proportionally equal to the values in 2010, though after taking into consideration the growth in the organic farming sector for cattle rearing. Data on the share of organic livestock was obtained from statistical data summarized by Eurostat at the national level [48]. For animals other than cattle, the share of organic production was disregarded since the share is very low (<1% of animals) [48]. Manure collection factors are given in Table A3 of Appendix A, for all animals and various types of housing.

2.3.3. Installed AD Capacity

Already installed AD capacity has to be considered when assessing additional potential implementation in the regions. Regional data on biogas installation was collected from various sources. In Veneto, a total of 220 biogas plants were in operation by 2018, of which 89% were considered agricultural plants, i.e., treating crop residues, energy crops and animal manures [49]. By contrast, 2566 plants were installed in Bavaria by 2019 [50], of which 93% were considered agricultural AD [7], while the rest were landfill gas and sewage gas. A breakdown of types of installed capacity (scale) was obtained from a census of installed plants [51] in 2011 in the Veneto region. It was assumed that installation continued in the same fashion through to 2018, with a preference for plants of capacity slightly lower than 1MW, due to an all-encompassing subsidy [52]. For Bavaria, data obtained was detailed down to city/rural district level, which made it possible to use average capacity to determine the scale breakdown of installed capacity. The types of capacity installed estimated for Veneto and Bavaria are shown in Table 4.

Table 4. Scale of installed biogas plants in Veneto and Bavaria.

Type of Capacity	Veneto 2018		Bavaria 2019	
(kWe)	n	%	n.	%
<100	23	12%	9	0%
101–500	43	22%	1352	56%
501–1000	118	60%	1010	42%
>1000	3	1%	11	0%
Biogas in broiler	0	0%	0	0%
No data	10	5%	15	1%

2.3.4. Regional Energetic Potential

The methane potential of various feedstocks (Table A4, Appendix A) was used to derive the quantities of feedstock currently being processed by the already installed AD capacity in each region. Since it was not possible to obtain specific data on precisely what types of feedstock are used at the NUTS2 level, statistics on the manure to crop share processed in AD were scaled down from national to regional level. For Germany, feedstock inputs for agricultural biogas plants are on average 45% manures and 55% crop material [28], while in Veneto the mix is on average 55% manures and 45% crop material [53]. A CHP electrical efficiency of 38% and a value of 9.97 kWh per liter of methane (CH_4) were assumed. The capacity installed in each region corresponds to 137 MW in Veneto and 1237 MW in Bavaria. Taking account for the installed capacity, the average mix of manure and crop material present in each region is then used to estimate more precisely the feedstock already used in AD. The final available potential can then be calculated by taking the total agricultural feedstock produced and subtracting competing applications for animals, soil organic matter and already installed capacity.

2.4. TEA Method

TEA of the different technologies, utilizing different feedstock mixes was carried out. Financing costs, maintenance and plant overhead costs, labor related costs, and feedstock costs were aspects considered for the TEA. For all scenarios, it was assumed that the AD plant has a productivity of 8760 hours per year.

The CapEx of the AD plants were estimated using a CapEx of M€ 4 for a 1 MW plant complete with AD, H_2S washer, and generator as a reference, which scales with a power of 2/3 to the electricity output [28,54]. The AD + Booster technology requires extra equipment for the separation and heat treatment, but it also reduces the required hydraulic retention time and therefore the required equipment size of the digester. Based on expert knowledge, it was assumed that regarding the CapEx these aspects equalize and therefore the CapEx of the AD + Booster scenarios is equal to that of the AD plants. The PHB production requires extra equipment for separation, filtration, selection, accumulation, and concentration. Based on expert knowledge, the CapEx for the AD + PHB scenarios was estimated to be 25% higher compared to that of the AD plant.

The financing costs were based on an amortization of the CapEx over 10 years with no interest. Maintenance, tax, insurance, rent, plant overhead, environmental charges, and royalties were assumed to be 10% of the CapEx per year [55,56].

The AD plants were assumed to have a high level of automation, thus, the labor related costs for a 1 MW plant are based on a 1 shift position. Assuming that an operator earns a salary of €18/h and including costs for supervision (+25%), direct salary overhead (+63%), and general plant overhead (+122%) [55], resulted in total labor related costs of k€ 500/y. For the 200 kW plant the labor related costs were divided by five, assuming farm personnel are available part-time. As the PHB production requires a number of extra unit operations and produces an extra product, the labor related costs were assumed to be 50% higher.

The feedstock costs including raw material, and handling and transportation costs are shown in Table 5. The costs for the different types of manure were estimated based on short distance transport costs of manure of €1/ton wet weight (WW) and thereby depend on the dry weight (DW) content of each feedstock. Grass and corn silage were assumed to be produced close to the AD plant and costs were estimated based on [57] and [58]. The costs for wheat straw, corn stover, and soybean straw were based on baling and transportation costs. The costs for vine shoots were based on harvesting and transportation costs. The costs for grape pomace, sugar beet pulp, and grain were based on [58].

Table 5. Feedstock costs in euro per dry weight.

Feedstock	Costs
Chicken manure	€5/ton DW
Cow manure	€9/ton DW
Pig manure	€18/ton DW
Grass silage	€100/ton DW
Corn silage	€120/ton DW
Wheat straw	€40/ton DW
Corn stover	€40/ton DW
Soybean straw	€40/ton DW
Vine shoots	€60/ton DW
Grape pomace	€150/ton DW
Sugar beet pulp	€150/ton DW
Grain	€200/ton DW

Based on the total costs, the break-even prices for electricity and crude PHB were calculated. In the scenarios in which crude PHB is produced, the break-even price of electricity is equal to the regular AD scenario. The break-even prices were compared to selling prices of electricity and PHB (Table 6). As in the AD + PHB scenarios a concentrated crude PHB is produced, extra required purification costs were included. For comparison between the economic performance of each scenario, the required subsidy, i.e., the difference between the selling prices and the break-even prices was calculated.

Table 6. Product selling prices.

Product	Specification	Price	Reference
Electricity	Germany	€0.042/kWh	[59–61]
	Italy	€0.058/kWh	[59–61]
Thermal energy	Germany	€0.025/kWh	[28]
PHB	Purified PHB	€3.6/kg	[15,62]
	Purification costs	€1.8/kg	[62]

2.5. LCA Method

LCA is a standardized methodology governed by international standards and guidelines [8]. Among these, the ILCD handbook offers detailed guidance on how to carry out LCAs in accordance with the definitions set out by the European guidelines [63]. Using this guidance, the study at hand is considered a situation A "micro-level decision support", since structural changes are not foreseen to occur in the background system, due to the small share of biogas in the overall context of renewable energy. Thus, average mixes were used for the background system and replacement of substituted products. Where co-products are produced, such as in the case of AD + PHB, system expansion is used. The same was done for heat, which is produced as a by-product when biogas is burned in a CHP unit. Though in the latter no credits were awarded in the Veneto region for the produced heat, since this is not yet valorized in Italy [51], apart from what is used for own consumption from operation of the plant. In Germany, the situation is slightly different, and thus, a credit was given to the co-produced heat at a rate of 0.52 kwh heat/kwh electricity, based on the amount of heat utilized at national level [28].

Residue feedstocks that are presently not typically valorized, apart from biogas production, come into the system burden free, since the burden of production was allocated solely to the main product. This is the case for animal manures, pomace and vine shoots. However, for energy crops, the full burden of production was taken into account, i.e., maize silage, grain and grass silages. For agricultural residues currently valorized in the market, such as sugar beet pulp, corn stover, and soybean straw, the burden of production was distributed by economic allocation, while for wheat straw an existing Ecoinvent process was used. The allocation key is shown in Table 7.

Table 7. Economic allocation key for crop by-products.

	%	% of	Reference
Corn stover	47	maize production	[64,65]
Sugar beet pulp	6	sugar production	[66]
Soybean straw	12	soybean production	[67]

In order to visualize the benefit of digesting manure, emissions from storing manure have been included in the assessment. A period of 50 days of manure storage, minus two weeks of unavoidable storing to account for losses and manure in housing units, is avoided by instead treating the manure with the technology scenarios. The quantity of avoided methane is directly proportional to the quantity of manure available in the region or the amount of manure is the feedstock mix. Values used for the calculation are included in Table A5 of Appendix A.

The product system modelling software OpenLCA [68] was used for the modelling and subsequent analysis, utilizing the Ecoinvent v3 database [69]. ReCiPE Hierarchist (H) [70] was chosen as the impact assessment method, and results were generated at midpoint and endpoint. The time horizon for calculation of impacts is 100 years from point of emission.

2.5.1. Plant Level

The functional unit (FU) at plant level is the treatment of 1 ton of feedstock of local characteristics, defined in Table 1 for each plant. Biogas is burned in a CHP, producing heat and electricity. Electricity substitutes the production mix corresponding to the geographical location of each biogas plant. Heat utilization was modeled as substituted district heat for the 200 kW Bavarian plant based on their data, while there is no heat utilization for the industrial size plant in Veneto. PHB production offsets average global thermoplastic production (Table A6, Appendix A).

2.5.2. Regional Level

The FU at NUTS2 level is the treatment of all the AD compatible feedstock defined through the mass flow analysis of available potential for each region (see Section 3.1). An energetic cut off of 1% was applied, so that feedstocks contributing less than 1% of total energetic potential of all feedstock in the region were left out. To simplify matters further, partly due to results from the TEA, the regional assessment was done for plants of industrial size, i.e., 1000 kW for both locations, processing a feedstock mix corresponding to the regional availability, which is defined in the regional feedstock availability assessment. Transport for the regional and plant level assessments was included as 1 km of feedstock transport, and other distances were tested in a sensitivity analysis.

A second sensitivity analysis was also included. The energy grid of each location was replaced with a theoretical future green energy mix, in order to observe the effect of changing energy grids through time. This follows best practices for including partially dynamic LCA in systems with a long service life [71].

2.6. Interpretation of Environmental Impacts

In order to interpret the results, several methods were used. Because of political importance as well as ease of understanding, GHG emissions were used as a proxy for environmental impacts in some discussion, though due to the potential issues with only using GHG emissions, e.g., burden shifting [72], other interpretation methods are were also used. In particular, two methods were used: the first is a monetization of environmental impacts based on endpoint damages [73] and the second uses a form of multiple criteria decision assessment called technique for order of preference by similarity to ideal solution (TOPSIS) [74], utilizing the implementation method ArgCW-LCA [75].

In the first of these two methods, monetization and ReCiPe endpoint damages [76,77] are used to calculate the external costs of the implementation of a given technology at a given scale or region. This was done through two methods. The first, for ecosystem damages, is based on budget constrained ability to pay, which is used to derive a valuation for species years (Species.Yr) gained or lost [78], as this is suggested as the least uncertain method for this valuation [79]. For that valuation, 65,000 USD_{2003} per Species.Yr was utilized. In order to evaluate the disability adjusted life year (DALY) loss or gain, a value from Dong et al., who assessed a number of different methods, was utilized [80]. The valuation derived in these different methods varies significantly, on the range of 1 to 2 orders of magnitude. Therefore, here we used the average of these values, 110,000 USD_{2003} per DALY, which is also in line with the value derived from budget constraint monetization [78], which again should have the least uncertainty. Since resource scarcity endpoint damages are already expressed in monetary terms, no further interpretation is necessary.

In the second method used for deriving a single score, based on the ArgCW-LCA method [75], ReCiPe midpoint environmental impacts [76] along with a valuation of required subsidy for profitability to represent the economic impacts were used as the input criteria for TOPSIS utilizing weighting based on what Sohn et al., describe as a context weighting factor (CWF) [75]. Per a suggestion from the ArgCW-LCA method, as there was no specific stakeholder group present, the stakeholder perspective element was omitted from the method application. For this application, normalization for an average European person year emission was used [81]. Thus, weighting of the environmental impacts is derived, as described in the ArgCW-LCA method, by taking an average of two values: the average of the normalized midpoint impacts for impact category ‘i’ amongst all assessed scenarios, and the difference of the minimum and maximum normalized impacts for impact category ‘i’ amongst all assessed scenarios. This accomplishes two things: (1) taking the average of the normalized impacts scales the importance of emissions of the system to status quo emissions and (2) taking the difference between the maximum and minimum normalized impacts is to scale relative to the ability for choosing amongst the available alternatives to cause significant change in status quo emissions. This was completed for all impact categories resulting in the CWF for the environmental impacts. Economic impacts were ascribed a range of weights relative to the sum of weighting given to environmental impacts ranging from 10%–90%. The system was also run using equal weights for all criteria as a point of comparison to the context weighted and the other single score results.

3. Results and Discussion

3.1. Regional Feedstock Availability and Potential Bioenergy Production

A complete table of the sustainable/technical feedstock potential is presented in Appendix B, Table A7, for Bavaria and Veneto. These amounts were used for the TEA-LCA as the regional feedstock mix, though with a 1% cut-off based on the energetic potential of the feedstock.

When graphed on a % wt basis (Figure 4), a relatively large proportion of production of energy crops is evident in Bavaria. Both regions are rich in cattle manure and have a noteworthy amount of swine manure. After energy crops, the most abundant residues are cereal straw for Bavaria and sugar beet straw and soybean residues for Veneto. The regions notably differ from each other, in particular with regard to the production of certain crops, for example sugar beet, soya and grapes. The grapes, represented by pomace, are much more prominent in the Veneto region.

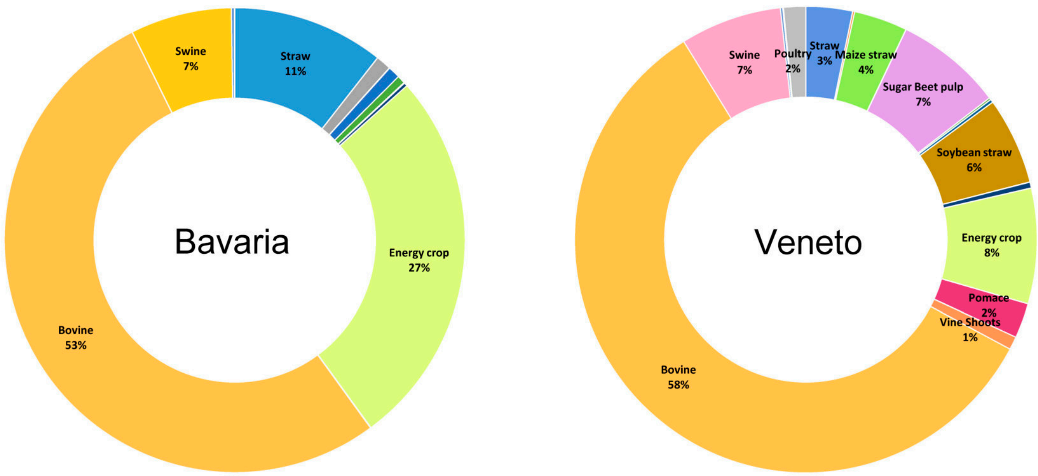

Figure 4. Share of total mix of agricultural residues on a % wt basis for Veneto and Bavaria.

In comparison to Veneto, Bavaria has a much larger share of energy crops, mainly maize silage. This greater share of energy crops is explained by feed incentives given to biogas plants using energy crops in Germany during 2004–2008 [82]. Although maize production has been capped by several German rulings, from 60% by mass input in 2014 lowered to 50% in 2018 and 44% by 2021, the combination of a high animal density and fodder production means that growing of maize has increased exponentially with unintended consequences, such as increasing land prices [22,82]. In Veneto, the feedstock mix exhibits more variability and the expansion of energy crops has not been as dramatic. This may likely be due to the Biogasdoneright™ concept promoted by the Italian Biogas Association, which originates in northern Italy, under which sequential crop cultivation is practiced, where the primary food crop goes to its intended purpose, and a secondary cover crop serves as feedstock for biogas plants [83].

Nevertheless, in energy terms, the potential of the feedstock mix is different than the availability based on mass, mostly due to the poor methane potential of some of the feedstocks. Without subtracting the feedstock that is already being used in the installed capacity of these regions, the energetic potential (based on electrical power) is seen in Figure 5. The largest share of potential is dominated by different feedstocks in the two regions. In Bavaria, the largest share can be obtained from energy crops, while in Veneto the largest share can come equally from cattle manure or energy crops. As a rough estimate, 153 PJ and 38 PJ remain as unexploited feedstock. This represents 31% and 54% of the total available feedstock potential, in Bavaria and Veneto, respectively, which is estimated as described in Section 2.3.4. However, for the LCA, all of the feedstock in the region was assumed to be utilized by the technologies, since in theory biogas plants can be retrofitted with the additional equipment needed for implementation of the AD + Booster technology and PHB production.

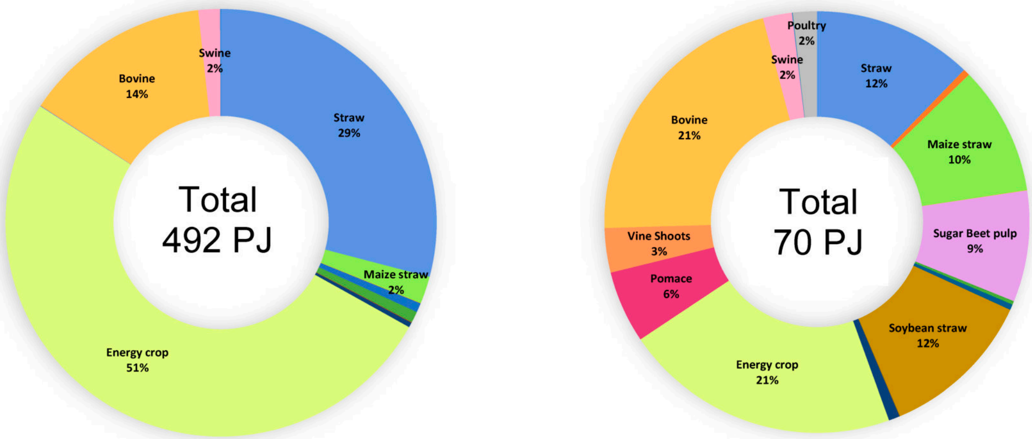

Figure 5. Energetic potential from agricultural residues for Bavaria (left) and Veneto (right) as % energy basis.

3.2. TEA Results

Based on the technical description of the different technologies and the different feedstock compositions, the flow sizes, flow compositions, production of electricity, heat, and crude PHB were estimated. Linking these process parameters to the economic parameters results in the TEA in Table 8.

In all scenarios, the financing, maintenance, labor-related, and feedstock costs are in the same order of magnitude. The contributions of these cost aspects to the total cost vary between 19% and 34%. The small-scale scenarios have, relative to annual production, a larger CapEx compared to the industrial scale, therefore financing and maintenance costs increase the break-even prices for the small-scale scenarios. This results in a break-even price are 34% higher for electricity and 27% higher for crude PHB for the small-scale scenarios, compared to the industrial scale scenarios. As all cost aspects are in the same order of magnitude, the extra required labor in the AD + PHB scenarios results in a significant contribution to the total costs. Logically, the extra labor related costs increase the break-even price of crude PHB. Compared to the feedstock costs of the studied plants, the regional level feedstock in both Bavaria and Veneto have a slightly higher contribution to the costs and to the break-even prices. In the Bavaria scenarios, the revenues of the thermal energy cause a reduction to the break-even prices of 8% for the small scale and 6% for the industrial scale, relative to scenarios that do not utilize the thermal energy.

For the 1 MW AD plant scenarios the average estimated break-even price for electricity is €0.22/kWh. For the AD + Booster scenarios, the average estimated break-even price for electricity is €0.19/kWh, a reduction of 12% in comparison to AD alone. Using the break-even for electricity of regular AD in the AD + PHB scenarios results in an estimated break-even price for crude PHB in the range €4.3/kg to €4.7/kg. When the purification costs of €1.8/kg are included, the break-even price range for PHB is in the range €6.1/kg to €6.5/kg. Due to the difference between market price and the break-even prices, as outlined in Section 2.4 (Table 6), it is clear that both electricity and PHB require large subsidy contributions to be profitably produced in AD plants. Relative to their respective market prices, the required amount of subsidy for the production of PHB is smaller compared to the subsidy for the production of electricity. Nevertheless, the production of PHB requires the co-production of electricity (Table 8).

Table 8. Techno-economic assessment (TEA) results of different scenarios.

Plant Level								
			Small scale (200 kW)			Industrial scale (1 MW)		
			AD	AD + Booster	AD + PHB	AD	AD + Booster	AD + PHB
CapEx		M€	1.4	1.4	1.4	4.0	4.0	4.0
Electricity	Produced	kW	200	224	138	1000	1124	662
	Internal use	kW	15	19	30	75	95	150
	Offset	kW	185	205	108	925	1029	512
Thermal energy	Produced	kW	326	365	224	1632	1834	1080
	Internal use	kW	131	146	90	653	734	432
	Offset	kW	196	219	135			
Crude PHB	Offset	ton/y			58			287
Costs	Financing	k€/y	137	137	171	400	400	500
	Maintenance, etc.	k€/y	137	137	171	400	400	500
	Labor-related	k€/y	100	100	150	500	500	750
	Feedstock	k€/y	142	142	142	440	440	440
	Total	k€/y	516	516	634	1740	1740	2190
Break-even price	Electricity	€/kWh	0.29	0.26	0.29	0.21	0.19	0.21
	Crude PHB	€/kg			5.7			4.3
Subsidy	Electricity	k€/y	405	393	236	1274	1222	705
	Crude PHB	k€/y			225			711
	Total	k€/y	405	393	460	1274	1222	1416
Regional Level								
			Bavaria region (1 MW)			Veneto region (1 MW)		
			AD	AD + Booster	AD + PHB	AD	AD + Booster	AD + PHB
CapEx		M€	4.0	4.0	5.0	4.0	4.0	5.0
Electricity	Produced	kW	1000	1144	742	1000	1155	755
	Internal use	kW	75	95	150	75	95	150
	Offset	kW	925	1049	592	925	1060	605
Thermal energy	Produced	kW	1632	1866	1211	1632	1885	1232
	Internal use	kW	653	746	485	653	754	493
	Offset	kW	481	545	308			
Crude PHB	Offset	ton/y			255			227
Costs	Financing	k€/y	400	400	500	400	400	500
	Maintenance, etc.	k€/y	400	400	500	400	400	500
	Labor-related	k€/y	500	500	750	500	500	750
	Feedstock	k€/y	558	558	558	509	509	509
	Total	k€/y	1858	1858	2308	1809	1809	2259
Break-even price	Electricity	€/kWh	0.22	0.19	0.22	0.22	0.19	0.22
	Crude PHB	€/kg			4.4			4.7
Subsidy	Electricity	k€/y	1415	1356	906	1343	1275	879
	Crude PHB	k€/y			660			666
	Total	k€/y	1415	1356	1567	1343	1275	1545

3.3. LCA Results

3.3.1. Midpoint Results

Results were obtained both at midpoint and endpoint level, using the ReCiPE 2016 (H) LCIA methodology. The results were internally normalized and ranked relative to the best-performing technology scenario. Midpoint level results for both regions and scales showed, for the most part, the same technology preference, pointing to AD + Booster as the best performer across impact categories (ICs), followed by AD and lastly AD + PHB. In the Veneto region, slightly more variation is observed across impact categories (Figure 6) and AD + PHB can at times be the best performer, as seen in the Ionizing Radiation, Land Use, and the Mineral Resource Scarcity ICs. The importance of this variation was tested with TOPSIS and is discussed further in Section 3.4.

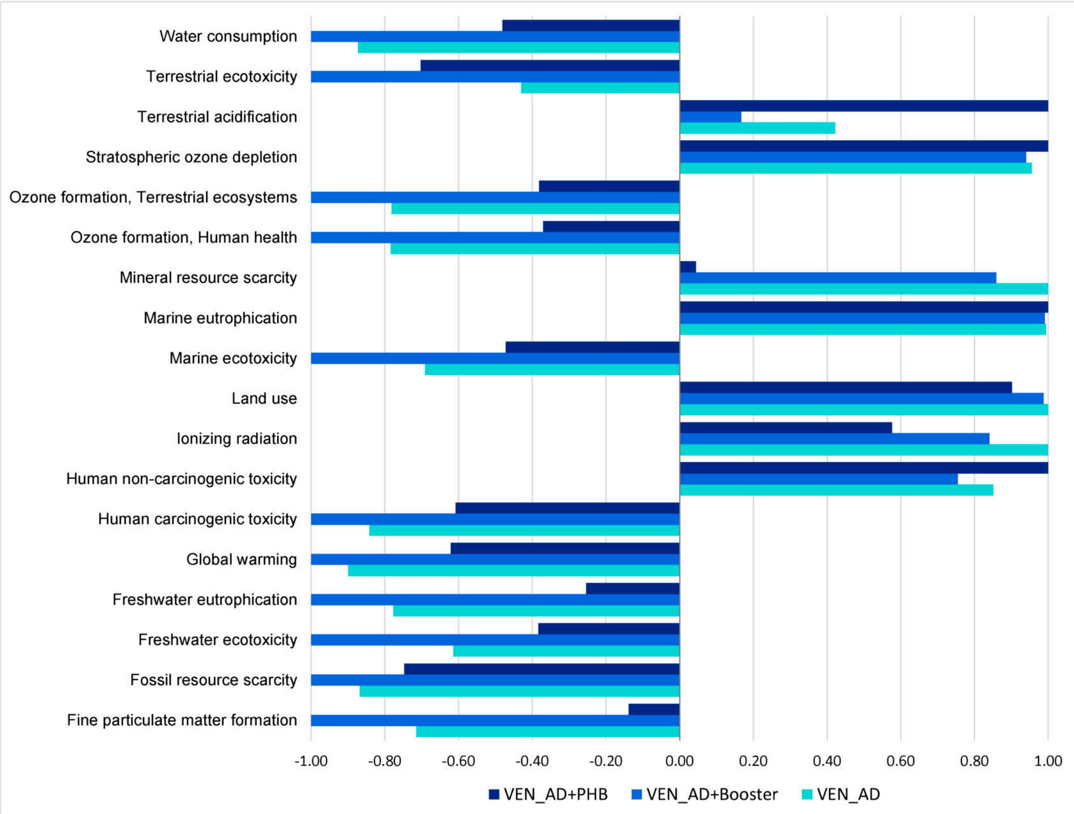

Figure 6. ReCiPE (H) Midpoint results for the region of Veneto for the three technology options i.e., AD, AD + Booster, and AD + PHB. Results are normalized per impact category to the worst or best performing scenario. Negative values show impact savings while positive values show burdens.

Midpoint results for the two farms assessed the small scale 200 kW farm in Bavaria and the 1000 kW farm in the Veneto region showed identical preference to the regional assessment when ranked within geographical location. However, more rank switching is observed when ranking is done across scales; this is explored further and discussed in Section 3.4., where rank reversal is checked thoroughly for both regional and scale assessments. Figures of normalized midpoint impacts for the Bavarian region, small and industrial scale are shown in Appendix B (Figures A1–A3), as well as tables of raw midpoint/endpoint results (Tables A8–A11).

3.3.2. Global Warming

As mentioned previously, global warming potential (GWP) shows the same technology preference as other ICs, with AD + Booster performing better than AD, which in turn performs better than AD + PHB. Looking at the contribution to GWP from the various elements that make up the system, it is possible to understand this preference. As can be seen in Figure 4, the higher energy production of the AD + Booster induces a higher electricity offset, which is largely responsible for the technology preference exhibited by the results. It is also evident that the offset for substituting plastic in the market for the AD + PHB options is very moderate and occurs on account of lower energy production, resulting overall in the lowest GWP savings out of all technology options. Figure 4 also shows the difference between the two regions on a per ton feedstock mix basis. An important difference can be observed in the crop mixture used for each region, where it is evident that Bavaria uses a more burdensome mix than Veneto. Other than crop differences, methane leaks from the facilities, here assumed to be 3% of the biogas produced, is an important source of GHGs. This is worth noting, as it can diminish the savings intended by these technologies. On the other hand, an important savings is attained by degassing animal manures, which would otherwise sit in storage facilities for a longer period producing methane that would be released to the atmosphere. This benefit can be seen in Figure 7 as the "methane offset storage" and is higher for Veneto due to the higher availability of animal manures on a %wt basis in this region.

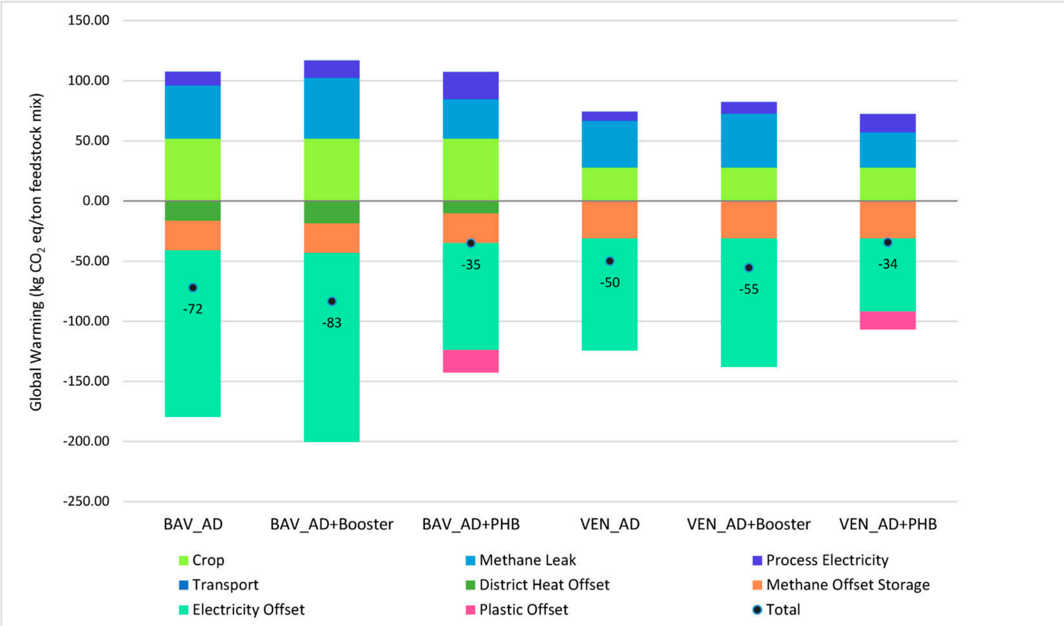

Figure 7. Global warming potential (GWP) contribution per ton of feedstock mix for the two regions, BAV for Bavaria and VEN for Veneto, for the three technology options, i.e., AD, AD + Booster, and AD + PHB.

Figure 8A,B, shows total GWP savings for both Veneto and Bavaria, respectively. As a total, the Bavaria region is capable of obtaining GWP savings 7.4, 7.7, and 5.4 times higher than in the Veneto region for AD, AD + Booster, and AD + PHB, respectively, on an annual basis. This is explained in part by the scale of the regions, feedstock density of the regions, as well as the energy density of each feedstock employed in the mix. While Veneto is also the smaller of the two regions, the lower GWP savings are partly due to an average 25% lower feedstock mass production per area relative to Bavaria. Moreover, the regional feedstock mix in Bavaria contains ca. 7% more crops and crop residues, among which maize silage is a prominent one, whilst Veneto contains ca. 7% more animal manures, which have a low methane/VFA productivity. The feedstock mix of Bavaria results in a higher electricity offsets, even though its feedstock mix contains a higher share of primary production (1st Generation) feedstock, i.e., maize silage, rather than secondary production such as straw. In addition, the utilization of waste heat in the Bavarian system for district heating gives an extra considerable impact offset to the region. If the heat were to be utilized in Veneto, then an extra 23%–25% savings in GWP could be attained there.

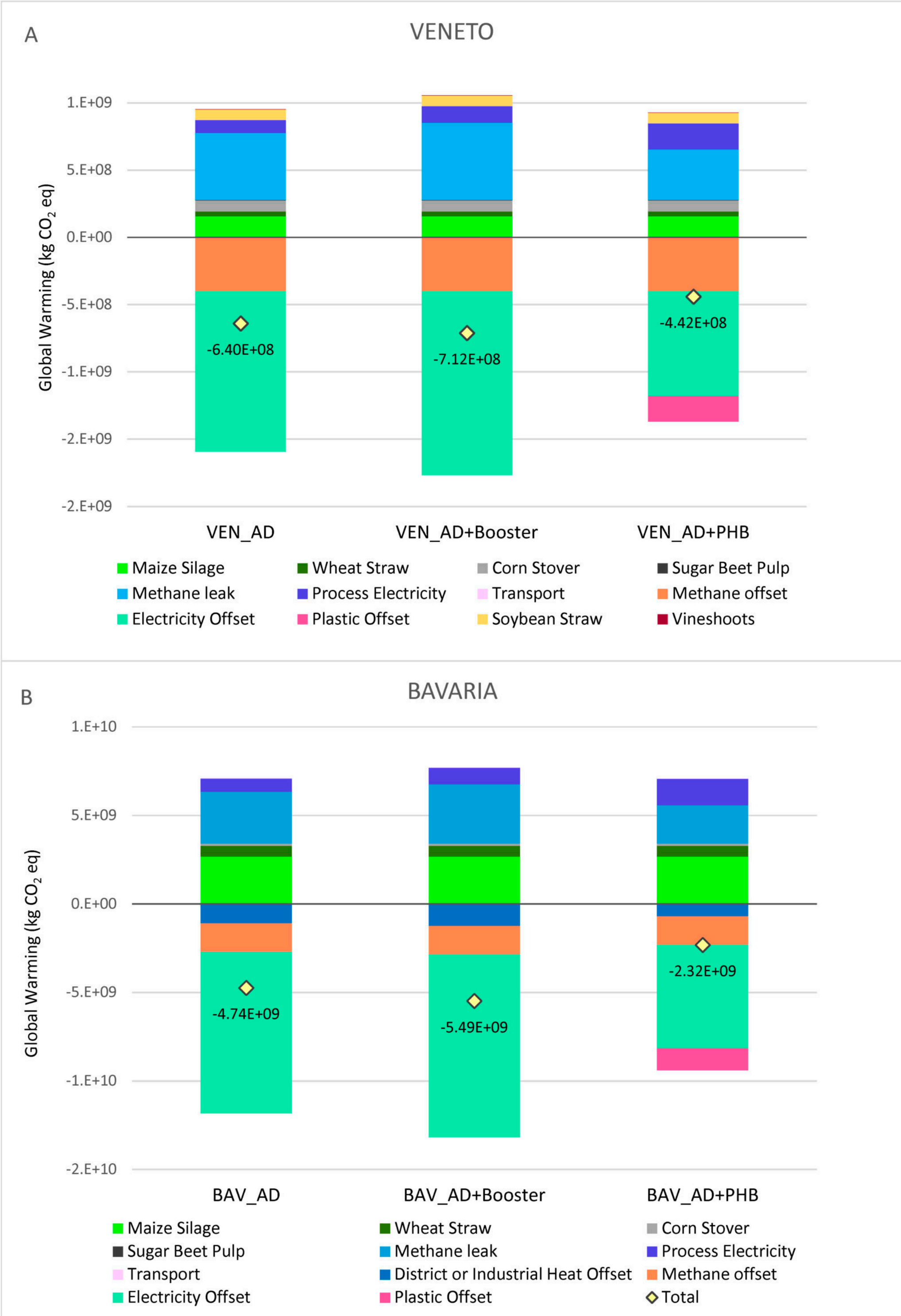

Figure 8. Global warming potential if all of the regional feedstock is treated on an annual basis for (**A**) Veneto and (**B**) Bavaria, as well as GWP contribution by the various system phases. Scenarios are named by the first three letters of the region (VEN or BAV) followed by each technology scenario (AD, AD + Booster, AD + PHB).

The pattern of feedstock efficiency is repeated when comparing the technologies on a scale basis. In fact, using more energy dense feedstock, i.e., feedstock that has a higher methane potential, leads to higher GWP savings for the small-scale facility (S + technology scenario), on a per ton feedstock basis, than for the industrial scale (I + technology scenario) (Figure 9). This is true even though the feedstock mix used in the small scale is more burdensome in terms of GWP, due to the cultivation phase of the feedstocks. The industrial scale facility still incurs savings to GWP, albeit lower, due to the poor characteristics of the feedstock utilized, which in this case is ca. 80% cow manure. Technology preference largely stays the same for both scales, though it is worth mentioning that a friendlier feedstock mix, i.e., with less first generation feedstocks, such as the one in the industrial scale is more important for the AD + PHB option, as can be observed when comparing S_AD + PHB and I_AD + PHB, which have savings of −15 and −25 kg CO_2 eq/ton, respectively.

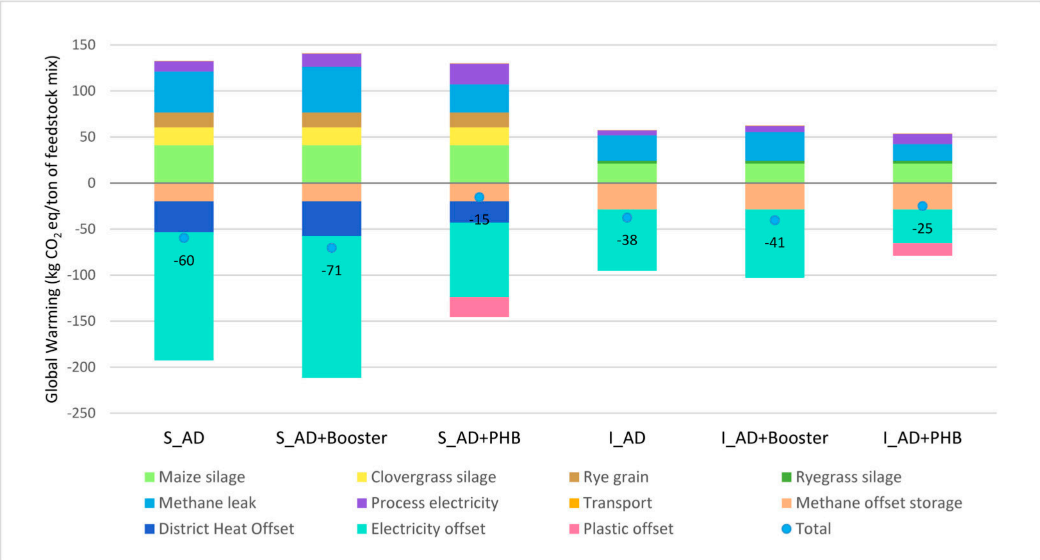

Figure 9. Global warming potential results for the small scale (200 kW) and industrial scale (1000 kW) cases, per ton of feedstock, as well as contribution to GW by each stage. Scenarios are named as S for small scale and I for industrial scale followed by each technology scenario (AD, AD + Booster, AD + PHB).

3.3.3. Sensitivity

Two parameters were tested to assess the sensitivity of the results: transport distance of the feedstock and the effect of a theoretical future green energy mix in the system.

The effect of transport varies depending on how well the technologies perform. The initial results, which include a 1 km transport distance were varied and transport was added up to 100 km. The result can be observed in Figure 10, where it is evident that a further transport distance can be allowed for the AD + Booster technology in both regions since this is the best performing technology. The point at which each technology scenario goes from GWP saving to GWP burden can also be seen in the graph. This point (the y-intercept) is 86, 99, and 42 kilometers respectively for BAV_AD, BAV_AD + Booster and BAV_AD + PHB, in Bavaria. In Veneto these distances are lower, because of the lower performance of the technologies in this region, where a transport distance below 59, 65, and 41 kilometers for VEN_AD, VEN_AD + Booster, and VEN_AD + PHB respectively, would ensure that the technologies continue to induce GWP savings. Needless to say, the lower the transport distances for the feedstock, the better the technologies perform.

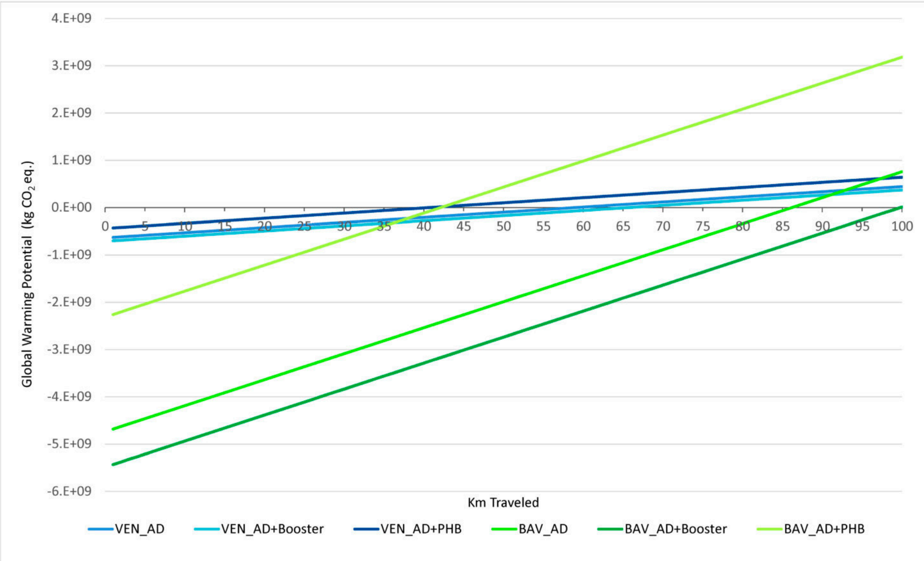

Figure 10. Effect of transport of feedstock on GWP savings. Scenarios are named by the first three letters of the region (VEN or BAV) followed by each technology scenario (AD, AD + Booster, AD + PHB).

The effect of switching the current production mix for the provisioning of process electricity and electricity offset with a future energy mix mainly composed of renewable sources is substantial for GWP results. For the regional assessment in Bavaria, all technology options result in impact burdens for GWP, while they continue to be impact savings for Veneto (Figure 11). This is due to the feedstock mix emissions in Bavaria, which are no longer counterbalanced by high emissions savings from offsetting of electricity. As has been shown before [26,84], offsets from replacing GHG intensive sources of electricity production such as coal, diminish as 'green' energy sources are implemented in the energy grid. The implications of this are very important for technologies producing renewable fuels, as their potential to produce savings will be bound to this future component.

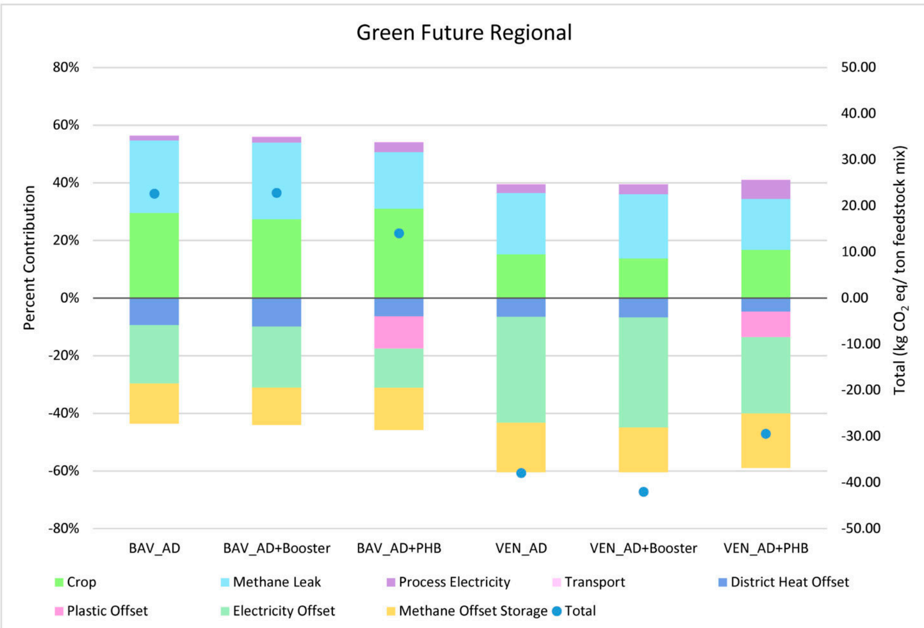

Figure 11. Global warming result for a future with a theoretical green energy mix. Scenarios are named by the first three letters of the region (VEN or BAV) followed by each technology scenario (AD, AD + Booster, AD + PHB).

On the other hand, BAV_AD + Booster, which is the worst performing scenario in terms of GWP continues to be the best performing scenario for most other impact categories in the green energy future SA (normalized midpoint results in Appendix B, Figures A4 and A5). As this clearly points to burden shifting the results were subjected to two single indicator interpretation methods to clarify the results.

3.4. Single Score Interpretations

Single score results, via TOPSIS developed by applying the ArgCW-LCA methodology [75], with environmental weights relating the results to a European's consumption patterns, and an economic weight derived from the TEA are discussed in this section. When assessed through TOPSIS (Figure 12), the initial regional results are very clear. Technology preference does not change within each region no matter which weighting is given to the results. AD + Booster is always the preferred choice, whether there are equal weights and high or low weight is given to economics. Furthermore, when impacts are monetized ($) so that the costs of environmental protection are visualized, these results also agree with the ArgCW-LCA and equal weights (EW) TOPSIS results. From the figure it is clear that the AD + Booster is also the best performer in terms of economic preference in Veneto (going up to 90% econ level), and on the contrary AD + PHB appears to be the worst. However, it is worth noting that in the Veneto region, if environmental concerns are weighed more heavily (<55% econ level), it is not easy to single out one of the technologies as unequivocally the best performing option, since the results perform close to equally well. This is not the case for Bavaria where the more burdensome feedstocks result in a more indisputable preference for the AD + Booster option, which produces the most energy. The implication of these results, namely that the more burdensome the energy production is, the more important the energy offsets become, is even more obvious for the plant level assessment. Here we see that though the technology preference is always the same (AD + Booster > AD > AD + PHB), the relative difference between the options becomes smaller the higher the economic weight (approaching 90%) for the Industrial plant in Veneto. This is a different pattern than the one observed for the regional level, where the distance between options, with and without PHB, increases with economic weight, and as supported by the assessment of midpoint results, the technology scenarios are closer to each other when the feedstock mix contains more animal manures than crop residues (see Figure 9). The same trend is seen for the small-scale plant in Bavaria, where the distance between the AD + Booster and AD + PHB option decreases with increasing economic weight. Though in this case, the plant's economic performance, which is very low in comparison to the industrial plant, is an important factor pulling all technology options further from the ideal.

The green energy future sensitivity was also checked with the single indicator methodology. The results again showed to be robust in terms of technology preference for the assessment (Figures A6 and A7). It is important to point out, however, that if the decision was based solely on GWP, then when looking at the green energy future one would choose AD + PHB in Bavaria, but continue to choose the AD + Booster in Veneto (Figure A6, Appendix B).

Overall the results are robust, though some clear patterns emerge. The single indicator results clearly highlight the dependency on the energy extraction efficiency of the options, which have increasing importance for regions with a more burdensome production, i.e., in the cultivation of energy crop for biogas production (the BAV and S scenarios). In this case, the electricity offsets are very important, not only for GWP, but all impact categories considered in an LCA, as evidenced by the single indicator preference. There are trade-offs when production utilizes a higher share of energy crops. On the one hand, electricity production is higher and with today's electricity mixes offsetting this type of production is highly valuable. On the other hand, it is worth noting that sustainability criteria for biofuels and biomass fuels might limit this type of production even more in the future. As it stands today, the renewable energy directive II sets out a cap on energy crops for renewable fuels and national caps are also present in various member states. The EC has also singled out feedstock of high potential for indirect land use change (iLUC), so that renewable fuels do provide the GHG reductions they are meant to bring. Though small plants are exempt from this cap (ca. <500 kW electric), one needs only to look at the German case, where around 50% of plants are small, as an example of how many small biogas plants can in fact have large consequences for how agricultural land is used.

The assessment also shows that varied production, i.e., not only energy, can be a viable option for plants with a high content of manures in the mix. In a future with an optimized PHB production this might be even more beneficial, also if we are to avoid the impacts of microplastic pollution, which are yet to be included in LCA studies. For now, strong subsidies are needed to increase technology penetration in the market with constant revision on sustainability targets. Continuing to green the energy grid should be a top priority by making as much energy as possible and fomenting technologies that increase the energy that can be obtained from biomass (like the AD booster). Future research on the possible synergies between technologies such as the AD-Booster + PHB could be interesting to explore.

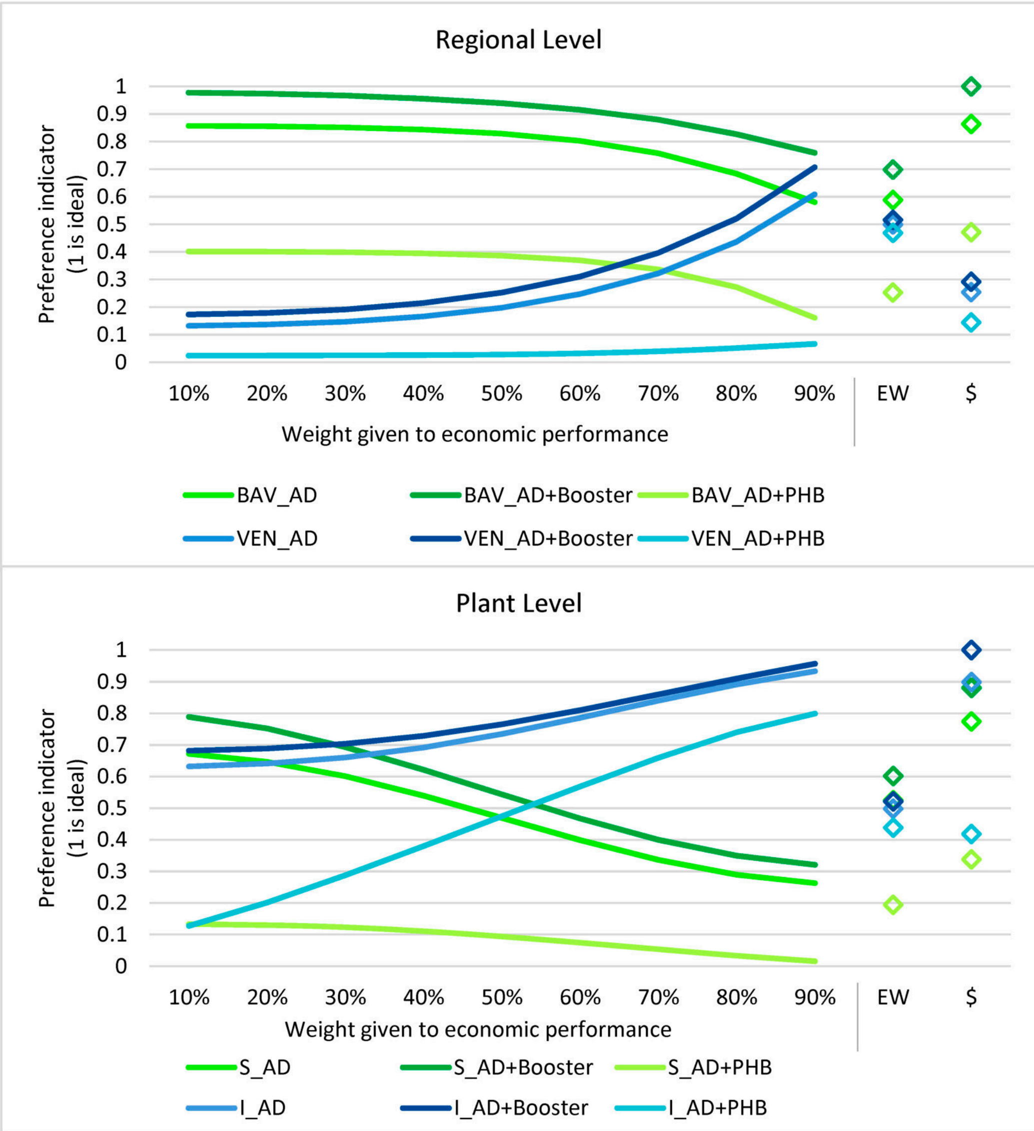

Figure 12. TOPSIS results for the regions (top) and scales (bottom), with varying economic importance (10% to 90%), equal weights (EW), and internally normalized monetization ($) of endpoint damages. Scenarios are named by the first three letters of the region (VEN or BAV) or scale size S for small and I for industrial, followed by each technology scenario (AD, AD + Booster, AD + PHB).

4. Conclusions

The production scale of the industrial set up assessed, with electricity ca. 1 MW and crude PHB production at ca. 300 ton/y, is small compared to their fossil and non-fossil alternatives. As a result, the financing, maintenance, and labor related costs increase the break-even prices significantly. Crude PHB production in AD plants requires the co-production of electricity in order to be adequately valorized, though benefits from avoided plastic particle pollution, which could be important, have not been included in the TEA and LCA. With today's energy mixes in the regions in question, it is highly valuable to offset electricity production and thereby options such as the AD + Booster are preferred for all environmental areas of protection. Material production in scenarios such as the AD + PHB perform equally well to more energy efficient scenarios for plants with a feedstock mix high in animal manures. Future caps on certain types of feedstock are worth considering when deciding on technology options to be implemented and/or subsidized.

Author Contributions: Conceptualization, G.C.V., J.V., J.S., and M.B.; methodology, G.C.V., J.V., and J.S.; formal analysis, G.C.V., J.V., and J.S.; writing—original draft preparation, G.C.V., J.V., J.S.; writing—review and editing, G.C.V., J.V., J.S., M.B., and S.I.O.; visualization, G.C.V., J.V., and J.S.; supervision, M.B. and S.I.O.; project administration, G.C.V.; funding acquisition, M.B. All authors have read and agreed to the published version of the manuscript.

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

Appendix A

Table A1. Grouping of crops, Eurostat names, and codes for crops and residue crop ratios.

Grouping	Eurostat Code and Name	Residue:Crop Ratio	Reference/Assumption for Residue:Crop Ratio
Cereal Straw	C1110-Common wheat and spelt	1.00	[19,21,85]
	C1111-Common winter wheat and spelt	1.00	assumed same as wheat
	C1120-Durum wheat	0.95	Assumed as triticale, [19,21,85]
	C1200 - Rye and winter cereal mixtures (maslin)	1.10	[19,21,85]
	C1300-Barley	0.93	[19,21,85]
	C1410-Oats	1.13	[19,21,85]
	C1420-Spring cereal mixtures (mixed grain other than maslin)	1.00	Average of common wheat, durum wheat, barley and rye
	C1600-Triticale	0.95	[19,21,85]
Rice Straw	C2000-Rice	1.70	[19,21,85]
Maize	C1500 - Grain maize and corn-cob-mix	1.13	[19,21,85]
Leguminous	P0000 - Dry pulses and protein crops for the production of grain (including seed and mixtures of cereals and pulses)	1.50	Assumed as soy
	P1100-Field peas	1.50	Assumed as soy
Oil-bearing	I1140-Linseed (oilflax)	1.42	[19,21,85]
Rape	I1110-Rape and turnip rape seeds	1.70	[19,21,85]
Sunflower	I1120-Sunflower seed	2.70	[19,21,85]
Soya	I1130-Soya	1.50	[19,21,85]
Industrial	I3000-Tobacco		Not relevant for regions
Energy Crop	C1700-Sorghum	1.30	[19,21,85]
	G3000-Green maize	1.00	Whole plant [21]
Forage	G1000-Temporary grasses and grazing	1.00	Whole plant [21]
	G2000-Leguminous plants harvested green	1.00	Whole plant [21]
	G9100-Other cereals harvested green (excluding green maize)	1.00	Whole plant [21]
Sugar Beet	R2000-Sugar beet (excluding seed)	0.23	[19,21,85]

Table A2. Livestock unit conversion factors and manure production per animal type [7].

	Livestock Unit	Manure	Manure
	LSU	kg/head/day	t/head/year
calves	0.40	8.00	2.90
bovine	0.70	20.00	7.30
male bovine	1.00	25.00	9.10
dairy cows	1.00	53.00	19.30
other cows	0.80	25.00	9.10
piglets	0.03	0.50	0.20
other pigs	0.30	4.50	1.60
sows	0.50	11.00	4.00
sheep	0.10	1.50	0.50
goat	0.10	1.50	0.50
broilers	0.01	0.10	0.04
laying hens	0.01	0.20	0.07
other poultry	0.03	0.30	0.11
Live poultry average	0.02	0.20	0.07

Table A3. Manure collectability factors based on different types of housing and type of production [47,48].

	Collectability
	factor
Stanchion	0.98
Loose housing	0.95
Organic	0.25
Poultry	0.98
Swine	0.98
Sheep	0.5
Goat	0.1

Table A4. Methane potentials of various feedstocks [7].

	DM	VS	Methane Yield	Methane Yield
	%	%	L CH4/kg VS	L CH4/kg fresh
Pig slurry	5.5	75	300	14
Cattle slurry	9	77.5	225	16.5
Poultry manure	20	75	325	52.5
Sheep [1]				16.5
Goat [1]				16.5
Maize silage [2]	35	92.5	350	119
Grass [3]	25	92.5	375	91.5
Alfalfa [4]	22.5	92.5	400	87.5
Sugar beet	17.5	92.5	305	51.5
Straw [5]	87.5	85	225	169
Pomace	35	92.5	600	194.5

[1] Assumed same as cattle slurry. [2] Used for energy crops. [3] Used for forage crops. [4] Used for leguminous crops. [5] Used for rice straw, rape straw, sunflower straw, soya straw, oil-bearing straw, industrial crop straw, and vine shoot.

Table A5. Parameters used for methane emission from manure storage [86].

		Cattle	Pig	Poultry
Dry matter content	kg DM/kg WW	10.8	5.5	20
Volatile solids	kg VS/kg DM	0.714	0.638	0.638
Methane production in storage (50 days)	g CH4/kg VS	19	98.5	98.5
Inevitable storage and losses (15 days)	g CH4/kg VS	5.7	29.55	29.55

Table A6. Composition of global average plastic production, including low density polyethylene (LDPE), high density polyethylene (HDPE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), polyethylene terephthalate (PET) and polylactic acid (PLA) [87].

Polymer Type	
LDPE	22.8%
HDPE	18.6%
PP	24.3%
PS	8.9%
PVC	13.6%
PET	11.8%
PLA	0.1%

Appendix B

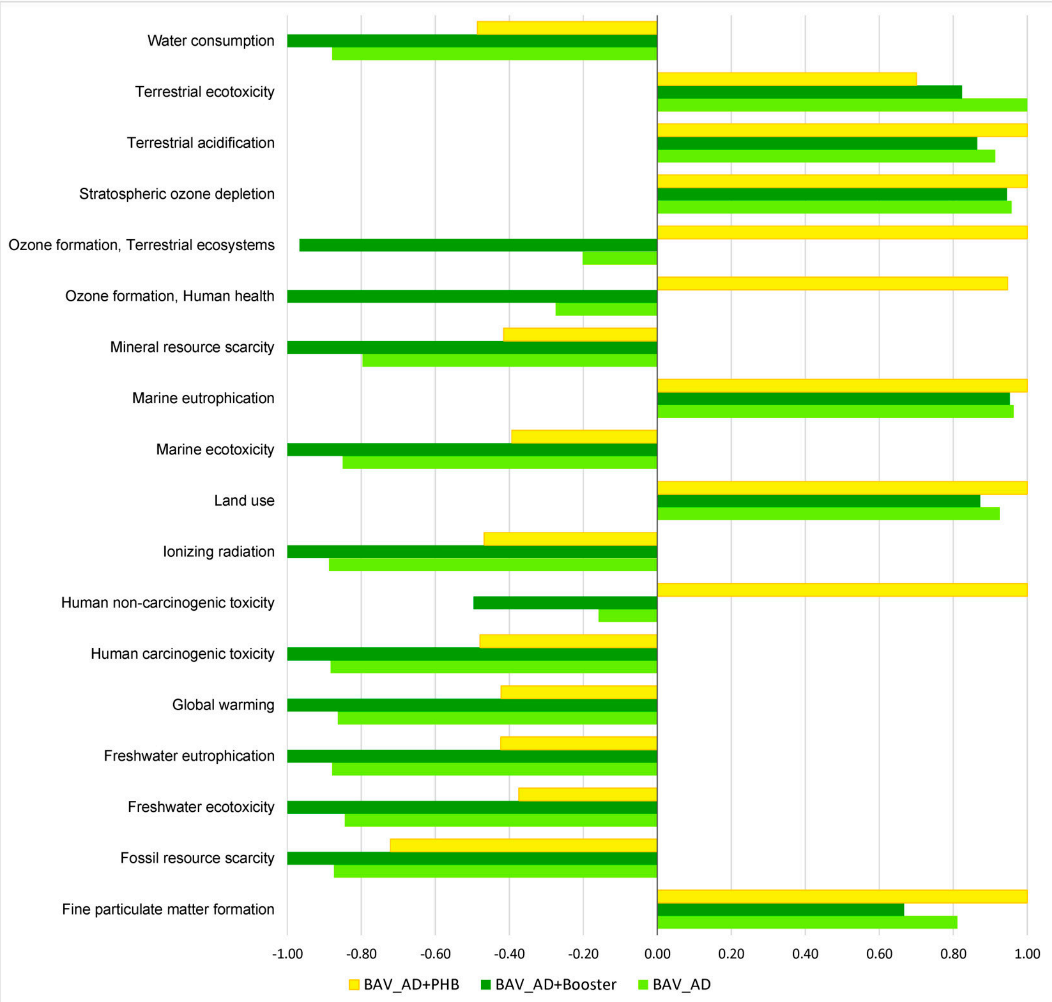

Figure A1. ReCiPE 2016 (H) midpoint results for the region of Bavaria. Results are normalized per impact category to the worst or best performing scenario. Negative values show impact savings while positive values show burdens.

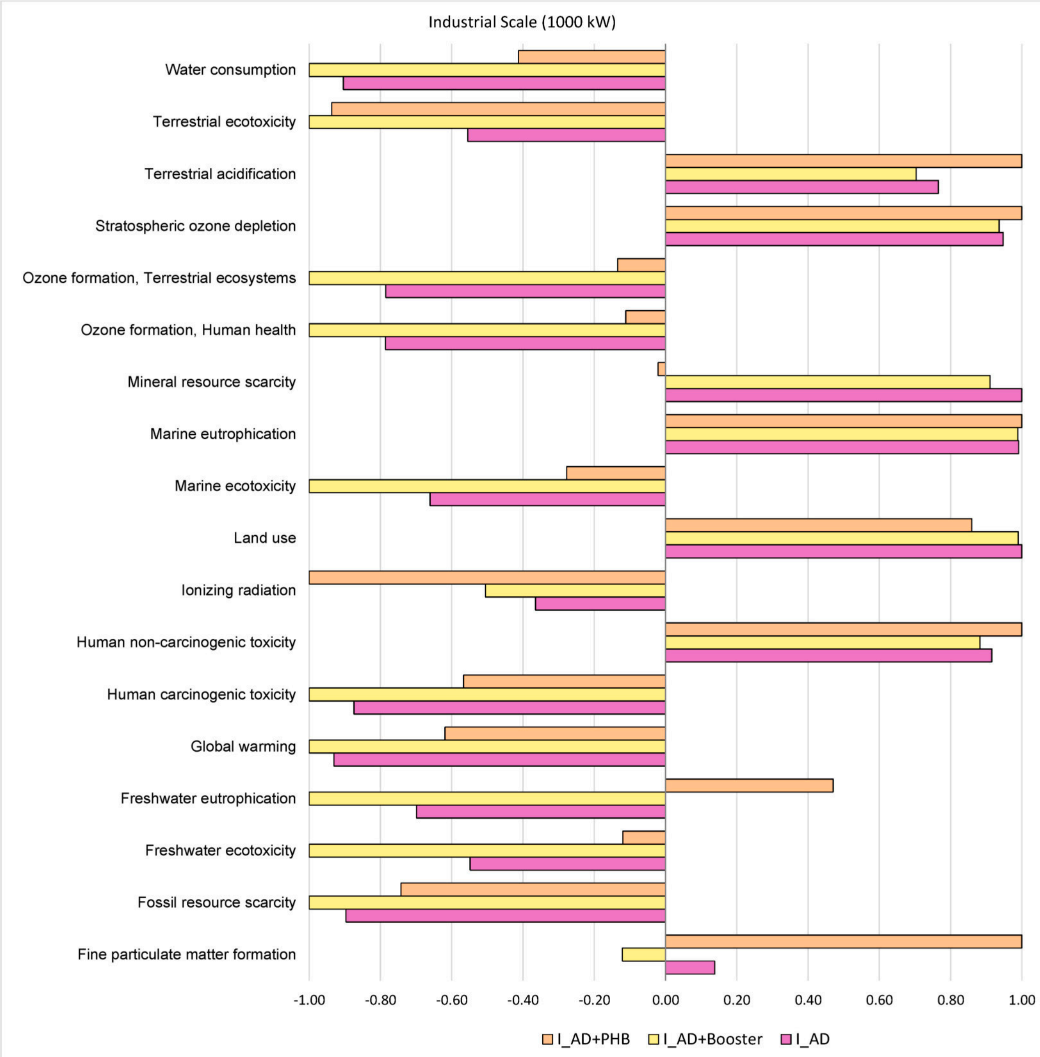

Figure A2. ReCiPE 2016 (H) midpoint results for the region of the industrial scale plant. Results are normalized per impact category to the worst or best performing scenario. Negative values show impact savings while positive values show burdens.

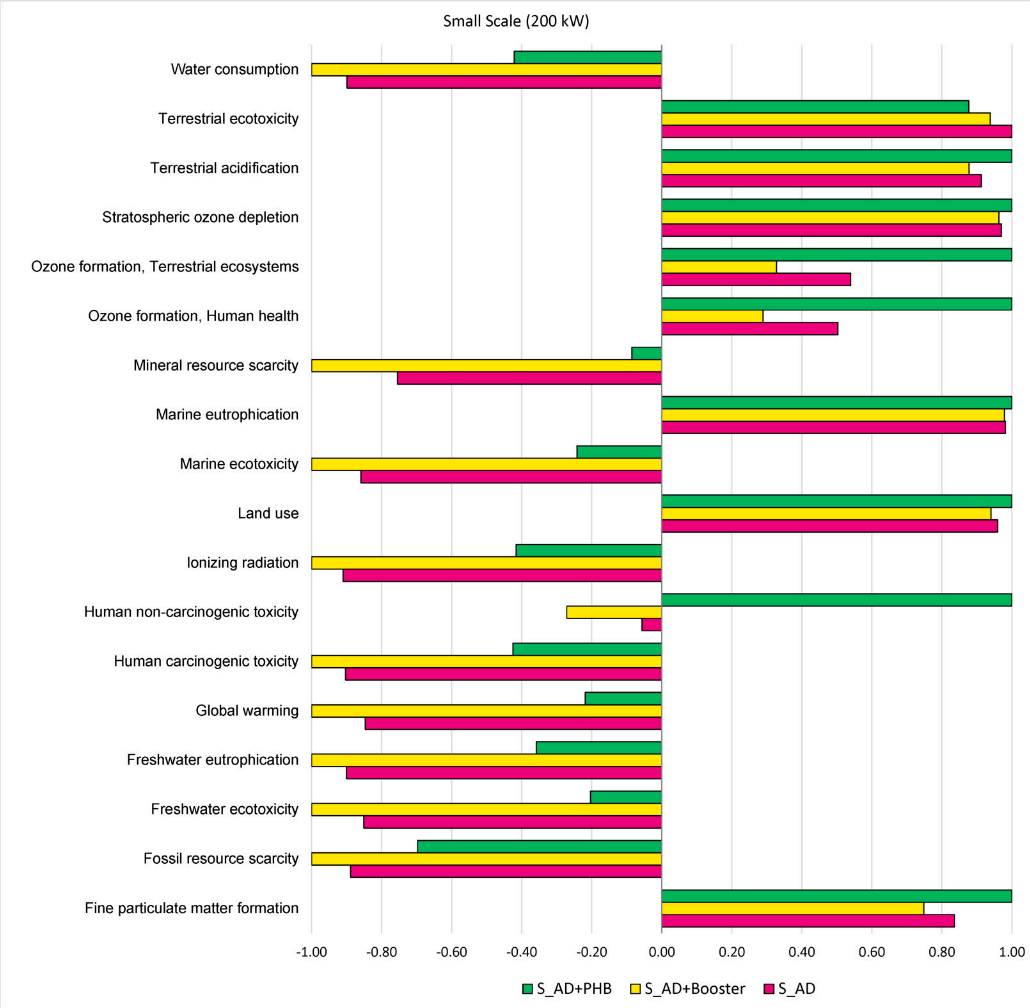

Figure A3. ReCiPE 2016 (H) midpoint results for the small-scale plant. Results are normalized per impact category to the worst or best performing scenario. Negative values show impact savings while positive values show burdens.

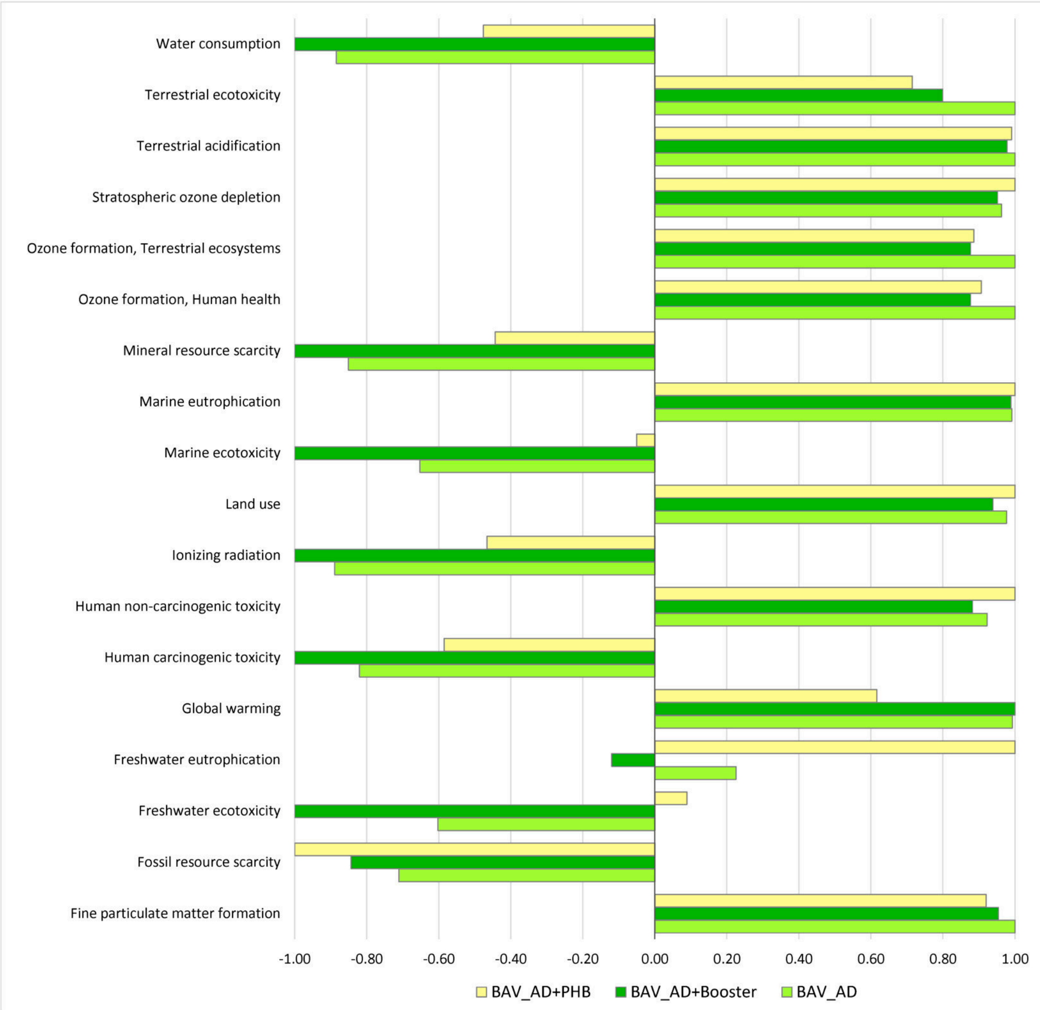

Figure A4. ReCiPE 2016 (H) midpoint results with theoretical green energy grid for the region of Bavaria. Results are normalized per impact category to the worst or best performing scenario. Negative values show impact savings while positive values show burdens.

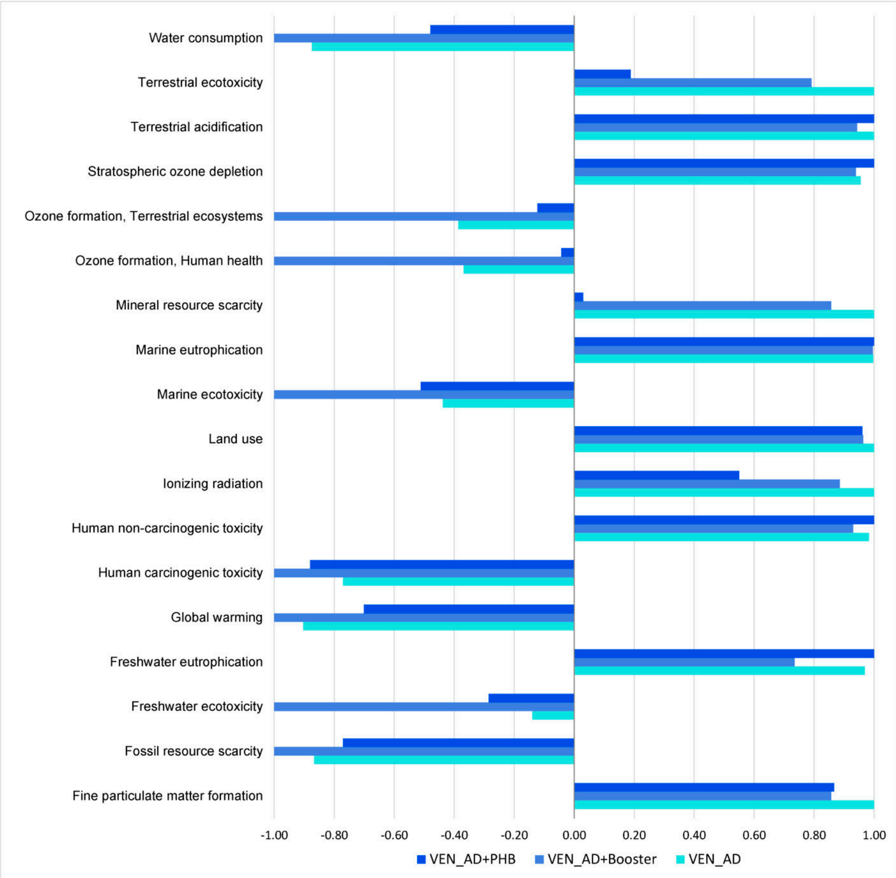

Figure A5. ReCiPE 2016 (H) midpoint results with theoretical green energy grid for the region of Veneto. Results are normalized per impact category to the worst or best performing scenario. Negative values show impact savings while positive values show burdens.

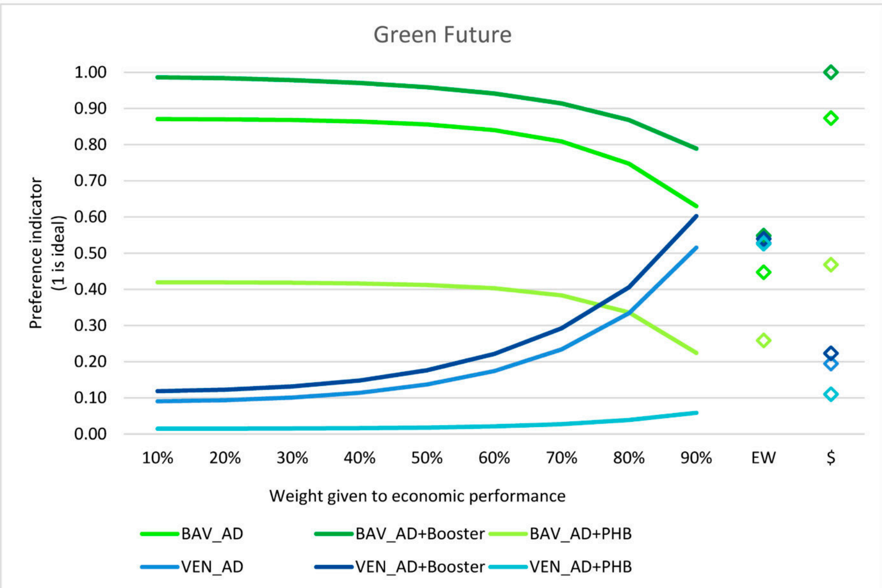

Figure A6. TOPSIS results for the regions with the theoretical green energy mix, with varying economic importance (10% to 90%), equal weights (EW), and internally normalized monetization ($) of endpoint damages.

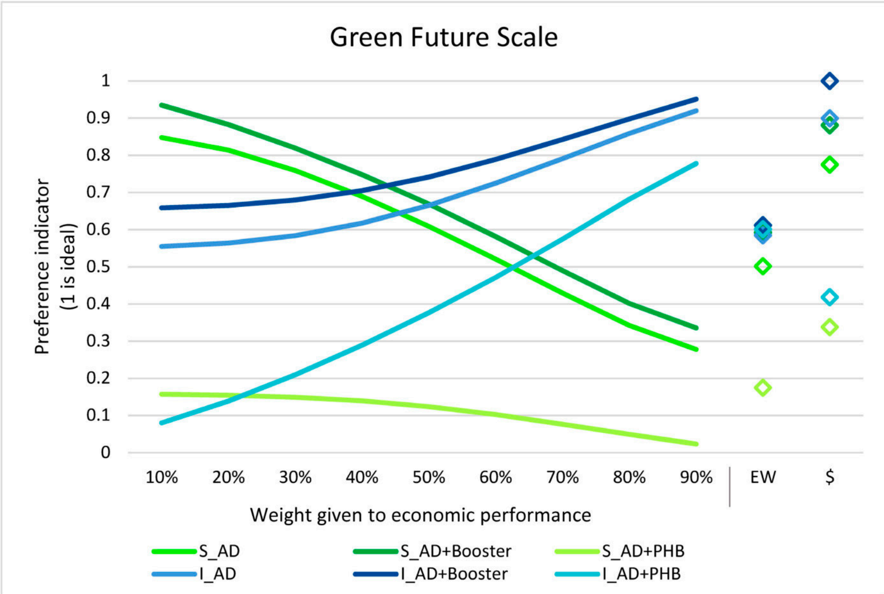

Figure A7. TOPSIS results for the two scales S = 200 kW and I 1000 kW, with the theoretical green energy mix, with varying economic importance (10% to 90%), equal weights (EW), and internally normalized monetization ($) of endpoint damages. Scenarios are named as S for small scale and I for industrial scale followed by each technology scenario (AD, AD + Booster, AD + PHB).

Table A7. Total amount of sustainable/technical feedstock potential in Mtonne/year, sorted from highest to lowest amount.

	Bavaria	**Veneto**
Cattle manure	35.08	7.58
Energy crop	17.65	1.04
Straw	7.09	0.42
Swine manure	4.73	0.92
Corn Stover	0.71	0.49
Sugar Beet	0.56	0.97
Rape	0.38	0.02
Forage	0.17	0.05
Sheep manure	0.14	0.02
Soybean straw	0.02	0.79
Pomace	0.02	0.31
Poultry manure	0.01	0.20
Leguminous residue	0.01	0.00
Vine shoots	0.01	0.12
Sunflower straw	0.01	0.03
Goat manure	0.01	0.01
Rice straw	0.00	0.02
Oil crop residue	0.00	1.12×10^{-5}
Industrial crop residue	0.00	2.26×10^{-3}

Table A8. ReCiPE 2016 (H) midpoint results for the regional assessment.

	Scenario Name						
Indicator	**BAV_AD**	**BAV_AD + Booster**	**BAV_AD + PHB**	**VEN_AD**	**VEN_AD + Booster**	**VEN_AD + PHB**	**Unit**
Fine particulate matter formation	3.27×10^6	2.69×10^6	4.04×10^6	-3.89×10^5	-5.44×10^5	-7.52×10^4	kg PM2.5 eq
Fossil resource scarcity	-2.06×10^9	-2.35×10^9	-1.70×10^9	-3.03×10^8	-3.49×10^8	-2.61×10^8	kg oil eq
Freshwater ecotoxicity	-2.54×10^8	-3.01×10^8	-1.13×10^8	-3.52×10^6	-5.72×10^6	-2.19×10^6	kg 1,4-DCB
Freshwater eutrophication	-1.12×10^7	-1.27×10^7	-5.39×10^6	-1.18×10^5	-1.52×10^5	-3.86×10^4	kg P eq
Global warming	-4.74×10^9	-5.49×10^9	-2.32×10^9	-6.40×10^8	-7.12×10^8	-4.42×10^8	kg CO2 eq
Human carcinogenic toxicity	-5.77×10^8	-6.54×10^8	-3.13×10^8	-1.48×10^7	-1.75×10^7	-1.07×10^7	kg 1,4-DCB
Human non-carcinogenic toxicity	-5.59×10^8	-1.75×10^9	3.52×10^9	4.02×10^8	3.56×10^8	4.72×10^8	kg 1,4-DCB
Ionizing radiation	-1.35×10^9	-1.53×10^9	-7.14×10^8	5.87×10^6	4.93×10^6	3.38×10^6	kBq Co-60 eq
Land use	1.21×10^7	1.14×10^7	1.31×10^7	1.95×10^6	1.93×10^6	1.76×10^6	m2a crop eq
Marine ecotoxicity	-3.62×10^8	-4.25×10^8	-1.67×10^8	-6.66×10^6	-9.65×10^6	-4.56×10^6	kg 1,4-DCB
Marine eutrophication	1.02×10^7	1.01×10^7	1.06×10^7	1.42×10^6	1.41×10^6	1.43×10^6	kg N eq
Mineral resource scarcity	-6.88×10^5	-8.63×10^5	-3.58×10^5	3.79×10^4	3.26×10^4	1.68×10^3	kg Cu eq
Ozone formation, Human health	-4.79×10^5	-1.75×10^6	1.65×10^6	-9.61×10^5	-1.23×10^6	-4.54×10^5	kg NOx eq
Ozone formation, Terrestrial ecosystems	-3.38×10^5	-1.62×10^6	1.68×10^6	-9.67×10^5	-1.24×10^6	-4.72×10^5	kg NOx eq
Stratospheric ozone depletion	4.86×10^4	4.79×10^4	5.07×10^4	5.93×10^3	5.83×10^3	6.21×10^3	kg CFC11 eq
Terrestrial acidification	3.26×10^7	3.09×10^7	3.57×10^7	7.66×10^5	3.04×10^5	1.82×10^6	kg SO2 eq
Terrestrial ecotoxicity	2.73×10^9	2.25×10^9	1.91×10^9	-7.79×10^7	-1.81×10^8	-1.27×10^8	kg 1,4-DCB
Water consumption	-2.27×10^{10}	-2.59×10^{10}	-1.26×10^{10}	-6.75×10^9	-7.73×10^9	-3.72×10^9	m^3

Table A9. ReCiPE 2016 (H) midpoint results for the scale assessment.

	Scenario Name						
Indicator	**S_AD**	**S_AD + Booster**	**S_AD + PHB**	**I_AD**	**I_AD + Booster**	**I_AD + PHB**	**Unit**
Fine particulate matter formation	0.08	0.07	0.09	0.00	0.00	0.03	kg PM2.5 eq
Fossil resource scarcity	−33.44	−37.61	−26.22	−16.66	−18.58	−13.79	kg oil eq
Freshwater ecotoxicity	−3.26	−3.83	−0.78	−0.11	−0.20	−0.02	kg 1,4-DCB
Freshwater eutrophication	−0.17	−0.19	−0.07	0.00	0.00	0.00	kg P eq
Global warming	−59.78	−70.58	−15.43	−37.69	−40.51	−25.06	kg CO2 eq
Human carcinogenic toxicity	−8.73	−9.67	−4.11	−0.80	−0.92	−0.52	kg 1,4-DCB
Human non-carcinogenic toxicity	−3.81	−18.33	67.59	52.53	50.63	57.37	kg 1,4-DCB
Ionizing radiation	−20.37	−22.38	−9.31	−0.10	−0.14	−0.28	kBq Co-60 eq
Land use	0.37	0.37	0.39	0.10	0.10	0.08	m2a crop eq
Marine ecotoxicity	−4.74	−5.52	−1.33	−0.24	−0.37	−0.10	kg 1,4-DCB
Marine eutrophication	0.37	0.37	0.38	0.07	0.07	0.07	kg N eq
Mineral resource scarcity	−0.01	−0.01	0.00	0.00	0.00	0.00	kg Cu eq
Ozone formation, Human health	0.04	0.02	0.08	−0.04	−0.05	−0.01	kg NOx eq
Ozone formation, Terrestrial ecosystems	0.05	0.03	0.08	−0.04	−0.05	−0.01	kg NOx eq
Stratospheric ozone depletion	0.00	0.00	0.00	0.00	0.00	0.00	kg CFC11 eq
Terrestrial acidification	0.61	0.59	0.67	0.24	0.22	0.31	kg SO2 eq
Terrestrial ecotoxicity	106.40	99.83	93.32	−5.38	−9.70	−9.08	kg 1,4-DCB
Water consumption	−331.10	−368.38	−155.31	−386.92	−428.04	−176.73	m^3

Table A10. ReCiPE 2016 (H) endpoint results for the regional assessment.

	Scenario Name						
Indicator	**BAV_AD**	**BAV_AD + Booster**	**BAV_AD + PHB**	**VEN_AD**	**VEN_AD + Booster**	**VEN_AD + PHB**	**Unit**
Fine particulate matter formation	2.06×10^{3}	1.70×10^{3}	2.54×10^{3}	-2.44×10^{2}	-3.41×10^{2}	-4.67×10^{1}	DALY
Fossil resource scarcity	-1.69×10^{8}	-2.06×10^{8}	-3.28×10^{8}	-8.80×10^{7}	-1.02×10^{8}	-8.53×10^{7}	USD2013
Freshwater ecotoxicity	-1.76×10^{-1}	-2.09×10^{-1}	-7.80×10^{-2}	-2.43×10^{-3}	-3.96×10^{-3}	-1.52×10^{-3}	species.yr
Freshwater eutrophication	-7.49×10^{10}	-8.53×10^{10}	-3.61×10^{10}	-7.90×10^{-2}	-1.02×10^{-1}	-2.57×10^{-2}	species.yr
Global warming, Freshwater ecosystems	-3.63×10^{-4}	-4.20×10^{-4}	-1.77×10^{-4}	-4.90×10^{-5}	-5.44×10^{-5}	-3.38×10^{-5}	species.yr
Global warming, Human health	-4.39×10^{3}	-5.09×10^{3}	-2.15×10^{3}	-5.94×10^{2}	-6.60×10^{2}	-4.10×10^{2}	DALY
Global warming, Terrestrial ecosystems	-1.33×10^{1}	-1.54×10^{1}	-6.49×10^{10}	-1.79×10^{10}	-1.99×10^{10}	-1.24×10^{10}	species.yr
Human carcinogenic toxicity	-1.92×10^{3}	-2.17×10^{3}	-1.04×10^{3}	-4.90×10^{1}	-5.82×10^{1}	-3.54×10^{1}	DALY
Human non-carcinogenic toxicity	-1.28×10^{2}	-4.00×10^{2}	8.03×10^{2}	9.16×10^{1}	8.13×10^{1}	1.08×10^{2}	DALY
Ionizing radiation	-1.15×10^{1}	-1.29×10^{1}	-6.05×10^{10}	4.98×10^{-2}	4.18×10^{-2}	2.87×10^{-2}	DALY
Land use	1.08×10^{-1}	1.01×10^{-1}	1.16×10^{-1}	1.73×10^{-2}	1.71×10^{-2}	1.56×10^{-2}	species.yr
Marine ecotoxicity	-3.80×10^{-2}	-4.47×10^{-2}	-1.76×10^{-2}	-7.00×10^{-4}	-1.01×10^{-3}	-4.79×10^{-4}	species.yr
Marine eutrophication	1.73×10^{-2}	1.71×10^{-2}	1.80×10^{-2}	2.41×10^{-3}	2.40×10^{-3}	2.42×10^{-3}	species.yr
Mineral resource scarcity	-1.59×10^{5}	-2.00×10^{5}	-8.29×10^{4}	8.78×10^{3}	7.54×10^{3}	3.88×10^{2}	USD2013
Ozone formation, Human health	-4.36×10^{-1}	-1.59×10^{10}	1.51×10^{10}	-8.74×10^{-1}	-1.12×10^{10}	-4.13×10^{-1}	DALY
Ozone formation, Terrestrial ecosystems	-4.36×10^{-2}	-2.09×10^{-1}	2.16×10^{-1}	-1.25×10^{-1}	-1.60×10^{-1}	-6.09×10^{-2}	species.yr
Stratospheric ozone depletion	2.58×10^{1}	2.54×10^{1}	2.69×10^{1}	3.15×10^{10}	3.10×10^{10}	3.29×10^{10}	DALY
Terrestrial acidification	6.92×10^{10}	6.55×10^{10}	7.57×10^{10}	1.63×10^{-1}	6.53×10^{-2}	3.86×10^{-1}	species.yr
Terrestrial ecotoxicity	3.12×10^{-2}	2.57×10^{-2}	2.18×10^{-2}	-8.80×10^{-4}	-2.06×10^{-3}	-1.45×10^{-3}	species.yr
Water consumption, Aquatic ecosystems	-1.37×10^{-2}	-1.56×10^{-2}	-7.61×10^{-3}	-4.08×10^{-3}	-4.67×10^{-3}	-2.25×10^{-3}	species.yr
Water consumption, Human health	-5.05×10^{4}	-5.75×10^{4}	-2.80×10^{4}	-1.50×10^{4}	-1.72×10^{4}	-8.26×10^{3}	DALY
Water consumption, Terrestrial ecosystem	-3.07×10^{2}	-3.50×10^{2}	-1.70×10^{2}	-9.11×10^{1}	-1.04×10^{2}	-5.02×10^{1}	species.yr

Table A11. ReCiPE 2016 (H) Endpoint results for the scale assessment.

	Scenario Name						
Indicator	**S_AD**	**S_AD + Booster**	**S_AD + PHB**	**I_AD**	**I_AD + Booster**	**I_AD + PHB**	**Unit**
Fine particulate matter formation	4.73×10^{-5}	4.24×10^{-5}	5.66×10^{-5}	2.24×10^{-6}	-1.85×10^{-6}	1.58×10^{-5}	DALY
Fossil resource scarcity	-2.77×10^{10}	-3.38×10^{10}	-5.28×10^{10}	-4.82×10^{10}	-5.39×10^{10}	-4.66×10^{10}	USD2013
Freshwater ecotoxicity	-2.26×10^{-9}	-2.65×10^{-9}	-5.40×10^{-10}	-7.75×10^{-11}	-1.41×10^{-10}	-1.68×10^{-11}	species.yr
Freshwater eutrophication	-1.12×10^{-7}	-1.25×10^{-7}	-4.47×10^{-8}	-2.19×10^{-9}	-3.14×10^{-9}	1.49×10^{-9}	species.yr
Global warming, Freshwater ecosystems	-4.57×10^{-12}	-5.40×10^{-12}	-1.18×10^{-12}	-2.88×10^{-12}	-3.10×10^{-12}	-1.92×10^{-12}	species.yr
Global warming, Human health	-5.54×10^{-5}	-6.54×10^{-5}	-1.42×10^{-5}	-3.50×10^{-5}	-3.76×10^{-5}	-2.33×10^{-5}	DALY
Global warming, Terrestrial ecosystems	-1.67×10^{-7}	-1.98×10^{-7}	-4.33×10^{-8}	-1.06×10^{-7}	-1.13×10^{-7}	-7.02×10^{-8}	species.yr
Human carcinogenic toxicity	-2.90×10^{-5}	-3.21×10^{-5}	-1.36×10^{-5}	-2.66×10^{-6}	-3.04×10^{-6}	-1.73×10^{-6}	DALY
Human non-carcinogenic toxicity	-8.74×10^{-7}	-4.18×10^{-6}	1.54×10^{-5}	1.20×10^{-5}	1.15×10^{-5}	1.31×10^{-5}	DALY
Ionizing radiation	-1.73×10^{-7}	-1.90×10^{-7}	-7.90×10^{-8}	-8.60×10^{-10}	-1.19×10^{-9}	-2.36×10^{-9}	DALY
Land use	3.31×10^{-9}	3.25×10^{-9}	3.45×10^{-9}	8.55×10^{-10}	8.46×10^{-10}	7.35×10^{-10}	species.yr
Marine ecotoxicity	-4.98×10^{-10}	-5.80×10^{-10}	-1.40×10^{-10}	-2.55×10^{-11}	-3.87×10^{-11}	-1.07×10^{-11}	species.yr
Marine eutrophication	6.31×10^{-10}	6.28×10^{-10}	6.42×10^{-10}	1.18×10^{-10}	1.18×10^{-10}	1.19×10^{-10}	species.yr
Mineral resource scarcity	-1.48×10^{-3}	-1.97×10^{-3}	-1.67×10^{-4}	5.79×10^{-4}	5.27×10^{-4}	-1.23×10^{-5}	USD2013
Ozone formation, Human health	3.77×10^{-8}	2.17×10^{-8}	7.49×10^{-8}	-3.71×10^{-8}	-4.72×10^{-8}	-5.28×10^{-9}	DALY
Ozone formation, Terrestrial ecosystems	5.87×10^{-9}	3.57×10^{-9}	1.09×10^{-8}	-5.31×10^{-9}	-6.77×10^{-9}	-9.10×10^{-10}	species.yr
Stratospheric ozone depletion	6.61×10^{-7}	6.57×10^{-7}	6.82×10^{-7}	1.84×10^{-7}	1.81×10^{-7}	1.94×10^{-7}	DALY
Terrestrial acidification	1.29×10^{-7}	1.24×10^{-7}	1.42×10^{-7}	5.03×10^{-8}	4.62×10^{-8}	6.57×10^{-8}	species.yr
Terrestrial ecotoxicity	1.21×10^{-9}	1.14×10^{-9}	1.07×10^{-9}	-6.10×10^{-11}	-1.10×10^{-10}	-1.04×10^{-10}	species.yr
Water consumption, Aquatic ecosystems	-2.00×10^{-10}	-2.23×10^{-10}	-9.38×10^{-11}	-2.34×10^{-10}	-2.59×10^{-10}	-1.07×10^{-10}	species.yr
Water consumption, Human health	-7.35×10^{-4}	-8.18×10^{-4}	-3.45×10^{-4}	-8.59×10^{-4}	-9.50×10^{-4}	-3.92×10^{-4}	DALY
Water consumption, Terrestrial ecosystem	-4.47×10^{-6}	-4.97×10^{-6}	-2.10×10^{-6}	-5.22×10^{-6}	-5.78×10^{-6}	-2.39×10^{-6}	species.yr

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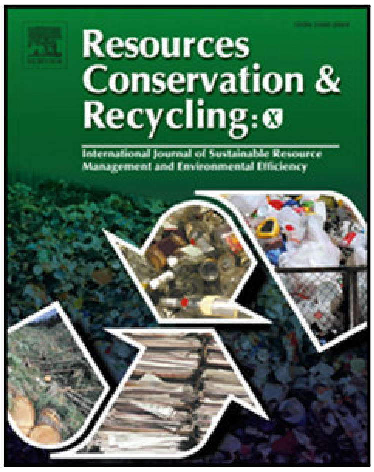

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Highlights

- Polyphenol extraction laboratory methods were improved through process design
- TEA and LCA were used to assess the designed industrial scale extraction
- TOPSIS-based MCDA was used to choose the best polyphenol extraction option
- Within feasible solvent ratios, SE exhibits better eco/enviro performance than PLE
- Preference for SE or PLE changes depending on level of importance assigned to economics

Insights from combining techno-economic and life cycle assessment – a case study of polyphenol extraction from red wine pomace

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Abstract

To determine the environmental and economic performance of emerging processes for the valorization of red wine pomace, a techno-economic assessment (TEA) and a life cycle assessment (LCA) are combined at an early design stage. A case study of two polyphenol extraction methods at laboratory scale, solvent extraction (SE) and pressurized liquid extraction (PLE), were first analyzed via a carbon footprint (CFP). Subsequently, the laboratory scale design was improved and translated into industrial scale and a TEA was performed on the industrial scale designs. Finally, LCA was applied again with all impact indicators and the information gathered from both the TEA and LCA was combined into concise decision support, using Multiple Criteria Decision Analysis (MCDA). SE performs better than PLE, due to a lower solvent to DW ratio and a less expensive processing setup in both environmental and economic terms. The CFP of at laboratory scale aided in showing potential environmental hotspots and highlighted the need to reduce solvent use. The MCDA showed a shift in decision support depending on how strongly economic or environmental benefits are valued and eases the interpretation of the 19 different indicators derived from the TEA-LCA results. Both SE and PLE with a solvent to dry weight (DW) ratio of 5 and 10, respectively, perform competitively while SE with a solvent to DW ratio of 10 outperforms PLE with a solvent to DW ratio of 25. The case study illustrated how early design calculations (CFP), and combined LCA and TEA may be combined to improve process design.

Key words: Techno-economic assessment, Life cycle assessment, polyphenol extraction, solvent extraction, pressurized liquid extraction

1. Introduction

Biomass demand for the production of bioenergy, biomaterials and biochemicals is estimated to increase by 70-110 % by 2050 compared to 2005 levels (Mauser et al., 2015). A paradigm shift to renewable sources of production has long been discussed, in the context of circular economy and valorization of biomass waste resources produced through the agricultural value chain. The bioeconomy today is estimated to have a €2.4 billion annual turnover, and it is only expected to increase in the future (Scarlat et al., 2015). Yet, the prefix bio does not guarantee sustainability. For example, growing biomass for biofuels has long been debated (Haberl et al., 2010; Murphy et al., 2011; Popp et al., 2014), prompting the Renewable Energy Directive (The European Commission, 2018) at an international, pan-European, level to ensure valid quantification of greenhouse gas reductions claims. In this regard, integration of methods such as life cycle assessment (LCA) and techno-economic assessment (TEA) are valuable input for quantitative sustainability assessments.

Combined TEA-LCA has been applied in many occasions to assess the environmental and economic ramifications of implementing new technologies (Cai et al., 2018; Hise et al., 2016; Vaskan et al., 2018). More interestingly, TEA-LCA has been used to quantify and monetize externalities, namely environmental damages, to provide a more complete picture of the financial burdens arising from environmental problems (Ögmundarson et al., 2018; Pizzol et al., 2015). Recently, combined TEA and LCA has been used to optimize new production routes from an early design phase, as in the case of integrated wastewater treatment and microalgae production for biodiesel production (Barlow et al., 2016), or the integration of power-to-gas technology of methane and photovoltaics (Collet et al., 2017). Combined TEA and LCA lends itself well to finding production hot spots and opportunities for optimization. This is even more relevant when applied to renewable resources such as biomass, which have to be managed sustainably.

New materials like biodegradable bio-sourced biopolymers and bioactive molecules such as, polyphenols obtained from agricultural residues can be combined to create new and innovative products (Vannini et al., 2019). Polyphenols present interesting possibilities as they can be utilized by various industries, such as in the pharmaceutical, nutraceutical and cosmetic industries (Pérez-López et al., 2014). Among other, polyphenols have been shown to have excellent health promoting qualities, such as anti-diabetic, anti-inflammatory, anti-bacterial and anti-cancer properties (Nowshehri et al., 2015). This versatility means that polyphenols may be used in niche markets as well as in mass markets, with various uses that may be of importance to the bioeconomy e.g. active packaging, coloring, food supplements, etc. Wine pomace is a residue rich in polyphenols, with a global production of 68 million tons of wine pomace annually (Nowshehri et al., 2015). To ensure a sustainable exploitation of polyphenol rich biomass, innovative polyphenol extraction methods at the laboratory scale were analyzed using TEA-LCA in order to identify hotspots and potentially environmentally problematic production steps.

On the other hand, results from the application of TEA-LCA can sometimes be confounding if, for example, one option performs better environmentally while incurring financial loss. The multitude of factors that must be taken into account remains an issue, when policy makers, corporations, or any other actor is faced with the need to decisively and definitively choose between alternative solutions to a given problem. In order to handle this, the decision-making context surrounding such a choice can be handled in many ways, from community-based decision making to round table discussions or even executive fiat. But, without a tool for interpreting fundamentally conflicting information, the results of the decision making process can vary wildly and may depend on happenstance and or subjective factors. Multiple Criteria Decision Analysis (MCDA) has been applied to aid in alleviating these problems by introducing a transparent and repeatable form of decision support (Kalbar and Das, 2020; Köksalan et al., 2011).

When assessing environmental issues in an LCA perspective, oftentimes practitioners turn to single indicators such as global warming potential (carbon foot-printing), but this poses potential downfalls such as burden shifting e.g. shifting environmental burdens from carbon emissions to environmental or human toxicity (Laurent et al., 2012). In other cases, practitioners turn to endpoint damage modeling, but these have high levels of uncertainty, can lead to unintentional bias (Kalbar et al., 2012a; Sohn et al., 2017), and still leave the decision maker with several categories of environmental damages e.g. ecosystem health, human heath, and resource availability. Furthermore, neither of these methods can be directly combined with economic indicators. In some cases, LCA practitioners have monetized impacts in order to combine environmental and economic indicators, however these suffer from issues, among others, involving the relationship of internalized and externalized costs (Reap et al., 2008). These issues have lead some LCA practitioners to turn to MCDA for providing decision support (Kalbar et al., 2016, 2012a; Sohn et al., 2017), as applying MCDA with a defined decision context to results from TEA-LCA is advantageous when a final decision must be taken.

Therefore, in this study LCA is applied at an early design stage to obtain a preliminary carbon footprint (CFP) of the polyphenol extraction methods. Subsequently, the design of the laboratory extraction procedures is improved and adapted to industrial scale and a TEA of the industrial scale scenarios is performed. Then LCA is applied again with all environmental indicators in simulated industrial conditions. This is done with the goal of obtaining a holistic picture of the economic feasibility and possible environmental impacts of each polyphenol extraction method. Lastly, MCDA is applied to the decision context of choosing between the polyphenol extraction methods and a weighting-profile derivation method (ArgCW-LCA) is applied (Sohn et al., 2020). The criteria from the LCA and TEA are incorporated to provide concise decision support for selecting one of the laboratory methods for scale-up.

2. Material and Methods

Results of laboratory scale experiments of different methods for the extraction of polyphenols from red wine pomace were evaluated using a combination of TEA and LCA. Two different labs, one located at the University of Bologna, Italy, and a second located at the Research Institute of Sweden (RISE), provided operational parameters for their laboratory setups. Yields, solvent amounts, temperature and time were then used to complete the inventory to carry out a preliminary carbon foot-printing (CFP) LCA of the laboratory scale experiments. The parameters of the most successful setups i.e. those producing the highest polyphenol yields, were used for the CFP and are described in detail in Table S1 of the supplementary information. The laboratory methods are described briefly in section 2.1. Following this step, industrial scale processes of the laboratory methods were designed and optimized for key parameters, using TEA (described in section 2.3). An LCA of the optimized industrial scale processes including all environmental indicators was then carried out. Lastly, a multiple criteria framework for decision support where the economic and environmental indicators are combined was applied to the results from the TEA-LCA.

2.1. Polyphenol extraction methods and laboratory experiments

The CFP of two different extraction methods, solvent extraction (SE) and pressurized liquid extraction (PLE), was determined. One SE setup and 3 different PLE setups, where the main difference is the solvent amounts used, were assessed for this step, of which the most successful setups in terms of yield are briefly described below, and the remainder can be found in the SI, since they did not become relevant for the industrial case. The laboratory extraction methods are also described in detail in (Ferri et al., 2020).

2.1.1. Solvent extraction with acetone

Solvent batch extraction was performed in the laboratory with various solvents (acetone, ethanol, and aqueous aceto-nitrile), temperatures (50 or 70°C), and extraction times (1, 2 or 4 hours). Of all operational parameters tested, an SE with the following conditions attained the highest polyphenol yield (Ferri et al., 2020). Solvent extraction with 61% acetone, and 39% water as solvent on a per mass basis, with a solvent to dry weight (DW) ratio of 11:1. Extraction was performed in an air-tight vessel at 50°C at atmospheric pressure where the solvent and previously ground pomace were kept in contact for 2 hours. Due to the polarity of polyphenols, they easily solubilize in polar media such as water/organic solvent and hydro-alcoholic mixtures. Once solubilized, polyphenols are carefully extracted from the liquid phase using a rotary evaporator under vacuum conditions, since many phenols also exhibit thermal instability. A powder is obtained from the rotary evaporatorwhich is then analyzed for polyphenol content of the extracts. Polyphenol content is expressed in kg gallic acid equivalents (kg GAE).

2.1.2. Pressurized liquid extraction with ethanol

As with SE, various operational conditions were tested for PLE. An ethanol/water (EtOH-H2O) mixture was used in combination with CO_2. The ratios of EtOH-H_2O:CO_2 varied from 75% to 50% and 100% in the various conditions tested, while the contact time tested varied from 30, 40 and 50 minutes (Ferri et al., 2020). PLE performed with 37% ethanol, 39% water and 25% supercritical CO_2 on a per mass basis was shown to attain the highest yield between the operational conditions tested. The extraction was performed at 80°C and 100 bar, at this temperature and pressure CO_2 is in the supercritical region, according to its phase diagram. As this is a continuous set up, where the solvent flows through the vessel containing the pomace, it leads to a high solvent to DW ratio of 101.

2.2. Carbon foot-printing of laboratory scale experiments

A CFP was performed on one SE and 3 PLE extraction methods, using only the Global Warming potential (GWP) impact category as the environmental indicator. The ReCiPe 2016 Midpoint Hierarchist (H) method (Huijbregts et al., 2017), which has a 100 year time horizon from point of emission, was used as impact assessment method, supplied by the Ecoinvent 3.4 Database (Wernet et al., 2016) and processed with the open source software OpenLCA (GreenDelta, 2019). The functional unit for the CFP is the production of 1 kg of polyphenols in kg GAE, assuming equal functionality. The process design software, SuperPro Designer v.10 (Intelligen Inc, 2018), was used to simulate the polyphenol extraction methods with industrial scale equipment. The operational parameters that attained the highest polyphenol yields in the laboratory (SE with acetone with a DW of 11:1, at 50 °C, for 2 hours and PLE with 75% EtOH:H2O, 25% supercritical CO_2 at 80 °C and 100 bar) were used for the CFP, as well as 2 other PLE shown only in the SI, Table S1. These operational parameters were used to populate the SuperPro Designer model so as to obtain the rest of the inventory of for example, energy and heat consumption, needed for the CFP. For the most part, the lab set up was kept the same. Through consultation with project partners it was possible to identify industrial scale equipment that would be able to perform the same functions as equipment in the lab, e.g. a spray dryer with nitrogen instead of a rotary evaporator for isolation of the polyphenols, distillation equipment for solvent recovery, etc.

The polyphenol producing plant is assumed to be placed in Italy and thereby, background processes for Italy from the Ecoinvent database were used as much as possible, e.g. the electricity grid.

2.3. Conceptual design of industrial scale processes

The process design focused on optimizing the operational parameters of the laboratory extraction methods so that it would be economically feasible to implement a polyphenol extraction at industrial scale. In order to achieve this, solvent recovery and product concentration are essential i.e. several process steps are required such as distillation, nano filtration, and spray drying (Figure 1 and Figure 2). The solvent loss and the energy required for solvent recovery should be reduced as much as possible. The solvent to DW ratio is an important parameter in solvent recovery. Industrial scale extraction processes usually have multiple extraction stages in a counter current flow setup (Berk, 2018). This setup reduces the required solvent to DW ratio and increases the product concentration in the extract, which reduces both the solvent recovery costs and the product concentration costs.

Based on literature (Dávila et al., 2017a, 2017b; Fiori, 2010; Todd and Baroutian, 2017; Viganó et al., 2017), process setup was designed for both SE (Figure 1) and PLE (Figure 2). Both designs assume multiple extraction stages in counter current flow. Compared to the laboratory scale experiments the residence times were adjusted as well as, flow and equipment sizes. The total extraction time is assumed to be 60 minutes for all processes. As shown in Ferri et al., (2020), the effect of lengthening extraction time was low on total polyphenol yields, thereby the yields obtained for 1 hour or 2 hours of extraction are comparable. This is why it was deemed possible to obtain the same polyphenol yields for SE even with a 60 minute residence time. Likewise, the authors found that a doubling of the acetone content for SE did not attain enough improvement of the polyphenol yield to justify the extra solvent use at industrial scale (Ferri et al., 2020).

A set up with counter current flow allows for a reduction of the solvent to DW ratio used in the laboratory scale experiments, while the extraction yield, i.e. the amount of polyphenols extracted per kg DW, is maintained. As mentioned previously the solvent to DW ratio is an important parameter. The reduction of the solvent to DW ratio in the industrial scale processes is difficult to estimate precisely, therefore, based on Dávila et al., (2017a, 2017b); Fiori, (2010); Todd and Baroutian, (2017); Viganó et al., (2017) and expert knowledge from the collaborating laboratories (Ferri et al., 2020), three feasible solvent to DW ratios were used in the TEA and LCA for each extraction method. The parameters of these scenarios are shown in Table 1. In all scenarios, the amount of polyphenols extracted is assumed to be equal to the laboratory scale experiments, since total residence times and solvent amounts are mostly within the ranges tested in the laboratory, though for a few of the scenarios it is important to validate the yields by further experiments i.e. SE-2 and, PLE-10 and PLE-25, which are assumed to attain the high yield due to the countercurrent set-up (Berk, 2018). The solvent to DW ratios and the solvent compositions were corrected for the amount of water in the pomace. The number in each scenario name refers to the solvent to DW ratio.

Table 1 Design parameters for industrial scale processes used in TEA and LCA.

	SE-10	SE-5	SE-2	PLE-50	PLE-25	PLE-10
Solvent to DW ratio (kg/kg DW)	10	5	2	50	25	10
Extraction stages	2	2	5		2	
Residence time (min/stage)	30	30	12		30	
Polyphenols extracted (g GAE/kg DW)		47			79	
Temperature (°C)		50			80	
Pressure (bar)		1			100	
Composition solvent						
- Water		33.3%			37.5%	
- Acetone		66.7%			-	
- Ethanol		-			37.5%	
- CO_2		-			25.0%	

The designs of both extraction processes include grinding of pomace to increase contact with the solvent, multiple extraction stages, distillation for solvent separation and recovery, nano filtration to concentrate the polyphenols, and finally spray drying for recovery of the polyphenols in powder form (Figure 1 and Figure 2). The solvent to DW ratio determines the concentration of polyphenols after extraction and distillation i.e. the higher the solvent use the lower the polyphenol concentration in the liquid. The extracted polyphenols after distillation are concentrated i.e. water is removed, by nano filtration to 25% DW and then to 95% DW by spray drying.

For SE, the solvent is recovered from the pomace by first pressing i.e. separating the majority of the solvent from the pomace and distilling the liquid fraction, while the pomace is sent to desolventizing (drying). The composition of the solvent in the recycle is 95% acetone and 5% water. For scenario SE-2, it is necessary to dry the pomace prior to extraction, because otherwise the required solvent composition cannot be obtained. This dryer is not shown in Figure 1, but is taken into account in the TEA and LCA.

For PLE, the solvent is recovered from the pomace by flashing the CO_2 and distilling the extract. The composition of the solvent in the recycle is assumed to be 90% ethanol and 10% water.

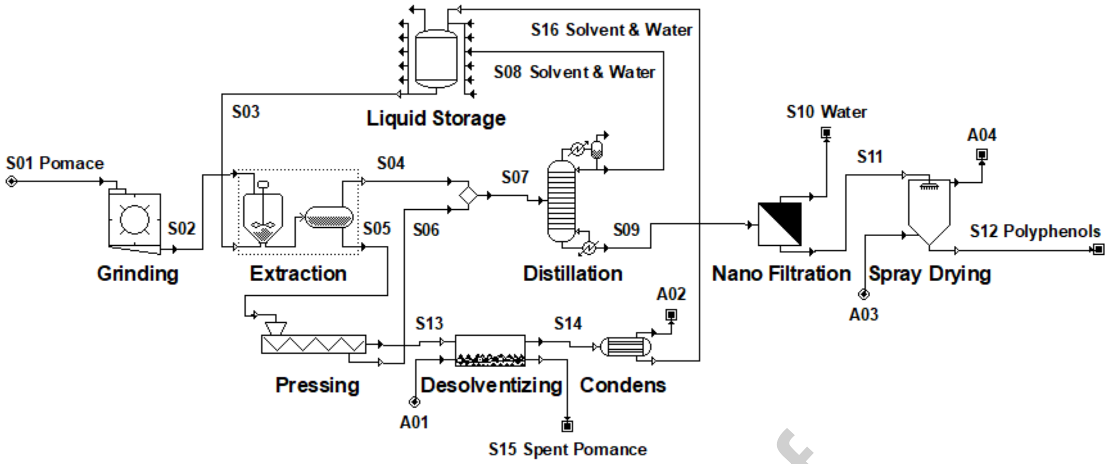

Figure 1 Process flow diagram for solvent extraction with acetone and water, for polyphenol recovery from grape pomace. Process includes input of wine pomace (S01), grinding, addition of solvents (S03) from liquid storage, extraction of polyphenols, distillation for solvent recovery and recycle (S08), nano filtration and spray drying for concentration and final recovery of polyphenols (S12), pressing and desolventizing of the wet pomace, condensation for additional recovery of solvent from the soaked pomace (S16).

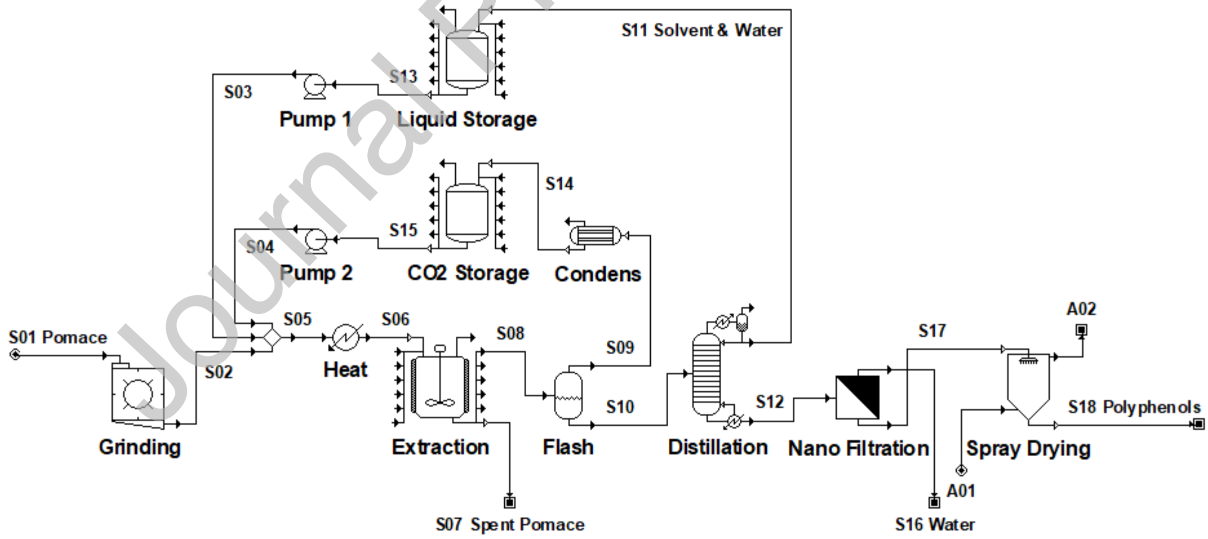

Figure 2 Process flow diagram for pressurized liquid extraction with ethanol, water, and supercritical CO_2 for the extraction of polyphenols from grape pomace. Process includes input of wine pomace (S01), grinding, pressurization by pump 1 and 2, addition of liquid solvents (S13) from liquid storage and supercritical CO_2 (S15) from CO_2 storage, extraction of polyphenols, flashing for CO2 recovery (S09) and distillation for liquid solvent recovery and recycle (S11),

nano filtration and spray drying for concentration and final recovery of polyphenols (S18). Spent pomace (S07) is not desolventized.

2.4. Techno-economic assessment of industrial scale processes

TEA of the designed industrial scale processes was carried out in order to investigate the economic repercussions of installing a polyphenol extracting plant. The TEA includes Capital Expenditure (CapEx) and Operating Expenditure (OpEx). Assumptions and simplifications were made in order to fill in data gaps. The most important assumptions considering the TEA are reported in Table 2. Assumptions of economic parameters were based on Intelligen Inc. (2018); Maroulis and Saravacos, (2007); Peters et al., (2003); and Sinnott and Towler, (2009).

Based on the flow sizes of the designed processes, equipment were scaled. Purchased equipment cost and CapEx were based on the literature used for the process designs (Dávila et al., 2017b, 2017a; Fiori, 2010; Todd and Baroutian, 2017; Viganó et al., 2017) and the references mentioned above. The CapEx of the extraction vessels was scaled using the six-tenths factor (Maroulis and Saravacos, 2007; Peters et al., 2003; and Sinnott and Towler, 2009). and was corrected for pressure (see detailed estimations in Table S2 of the SI).

In several wine growing areas wine pomaces and other residues are currently processed on industrial scale in centralized processing plants, so called distilleries. It is assumed that the polyphenol extraction will be performed in a setting similar to that of existing distilleries e.g. as in Italy and France, where 100% and 90% of wine pomace is sent to distilleries for treatment, respectively (Galanakis, 2017). The raw material costs for the polyphenol extraction are assumed to be negligible, since pomace is already part the current residue processing system.

The labor related costs were assumed to be the same for all scenarios and were based on: 2 shift positions, 4.8 operators per shift position, and an operator salary of k€ 30/y. Costs for supervision, direct salary overhead, and general plant overhead are added to the costs for operating labor.

Maintenance, including tax, insurance, rent, plant overhead, environmental charges, and royalties are assumed to be 10% of the CapEx per year. The financing costs are based on an amortization of the CapEx over 10 years with no interest (Peters et al., 2003; Sinnott and Towler, 2009).

Table 2 Parameters for the techno-economic assessment.

Production hours	8000	h/y
Red wine pomace	20	kton wet/y
	2500	kg wet/h
	36%	DW
Polyphenols extracted		
- with SE	340	ton GAE/y
- with PLE	572	ton GAE/y
Labor related costs	891	k€/y
Maintenance, etc.	10%	of CapEx/y
Financing costs	10%	of CapEx/y

The heat and electricity required in the different processes was based on process simulations in SuperPro Designer and on process parameters described in Maroulis and Saravacos, (2007); Peters et al., (2003); and Sinnott and Towler, (2009). Utility consumption was generalized to facilitate the techno-economic evaluation, thereby the heat required in the dryer for SE-2 and in the spray dryer, as well as the energy required for solvent recycle is assumed to be two times the heat of evaporation of the concerning stream, based on process simulations with the flow sizes of the designed industrial scale processes. For SE, this energy is distributed as follows: 90% for distillation (heat) and 10% for desolventizing (heat). For PLE, this energy is distributed as follows: 90% for distillation (heat), 5% for pumping (electricity), and 5% for heating prior to extraction. The electricity usage of the processing units is assumed to be: 10 kWh/ton input for grinding, 5 kWh/ton input for pressing, 5 kWh/ton permeate for nano filtration, 10 kWh/ton input for spray drying (atomization). Cooling water is used for cooling, for which the costs are assumed to be negligible. Despite all measures in the designed processes to recover the solvent, solvent loss is inevitable. Therefore, for all scenarios, a solvent loss of 2% of the solvent in the recycle is assumed. Prices, CO_2-equivalents, and heat of evaporation of relevant utilities and solvents are given in Table 3.

Table 3 Parameters for utilities and solvents

		Price €/kWh	GWP CO_2-eq/kWh
Electricity		0.10	0.43
Heat		0.04	0.37
Cooling		0.00	0.56
	ΔH vap kJ/kg	Price €/kg	GWP CO_2-eq/kg
Water	2260	0.00	0.0002
Ethanol	841	0.80	1.34
Acetone	539	1.20	2.87
CO_2	380	0.50	0.85

2.5. Life cycle assessment of industrial scale processes

Following the TEA, an accounting LCA was performed on the newly designed industrial systems as modelled by the TEA. The functional unit is the production of 1 kg of polyphenols expressed as 1 kg GAE. The assessment is a “gate-to-gate” LCA and includes all actions carried out in order to obtain polyphenols from red wine pomace. This includes all steps from when the pomace enters the production system to the product, the polyphenols, leaving the production facility, e.g. all processing steps, such as grinding, drying, adding solvents, filtering, distillation and more (Figure 1 and Figure 2). The assessment does not include the end of life of the polyphenols or any transport throughout the life cycle, since this is deemed equal for all processing methods. Also, any potential burden of the raw material, the red wine pomace, is not accounted for, since the wine pomace is waste from wine production. Likewise, no credits are assigned for the production of polyphenols potentially replacing similar products in the market. The LCA includes all 18 impact categories in ReCiPe 2016 Midpoint (H) methodology (Huijbregts et al., 2017). The geographical location of the polyphenol plant is Italy.

2.6. Development of weighting for Multi-Criteria Decision Analysis

In order to incorporate the various environmental, as well as the economic criteria derived from the results from the previously described TEA and LCA assessments (see section 3.2 and 3.3), the Technique for Order of Preference by Similarity to Ideal Solution (TOPSIS) method of MCDA (Hwang and Yoon, 1981) is used. This method applies compensatory aggregation based on the definition of a positive ideal solution and a negative ideal solution, a theoretical best and worst case scenario respectively, and selecting the alternative with the shortest geometric distance from the positive ideal solution and the longest geometric distance from the negative ideal solution after weighting is applied for each criterion. This method of MCDA is chosen due to its previous application in the context of LCA and because it is one of the most widely applied compensatory methods of MCDA when cardinal indicators are available for all alternatives (Kalbar et al., 2012b; Kalbar and Das, 2020).

All midpoint indicators from LCA and production prices of the various polyphenol production methods from the TEA (Table S3) are used as criteria in the application of TOPSIS.

When applying TOPSIS, there is an inherent application of weighting, even in its default mode, equal weights are applied (Pizzol et al., 2017). This presents a problem because the selection of the ideal alternative is directly related to weighting, which is further discussed in section 4.1.1. In this case, following the ArgCW-LCA method (Sohn et al., 2020), normalization factors (NF) (PRé, 2019) per impact category (i) are used to derive a relative importance factor (RIF), relating the average value, amongst all of the alternative extraction methods, of each of the midpoint impacts (MI) to the average European's annual environmental impact such that $RIF_i = \overline{MI_i} \,/\, NF_i$ represents the relationship between environmental and other criteria (Equation 1). For example, for calculating the RIF_{GW}, if the average GW impact amongst all assessed technologies ($\overline{MI_{GW}}$), were 80 kg CO_2 eq., then because the NF_{GW} for GW is 7990 kg CO_2 eq., the RIF_{GW} will be approximately equal to 0.01. In this case, production cost is then normalized such that production cost is allocated the desired weight and the sum of all weights is equal to 1000. The resultant weighting is then displayed in tabular form to promote full transparency in the assessment (Table S4, and Table S5).

3. Results and Discussion

3.1. Carbon foot-printing of laboratory scale experiments

The CFP analysis clearly shows that if laboratory conditions are maintained at industrial scale, then the acetone based solvent extraction method outperforms all other scenarios by a large margin, in terms of global warming potential (GWP), Figure 3. This is largely due to the amounts of solvent used in each scenario, which are lowest for the Lab-SE-11 scenario. The large amount of solvent used in the continuous setup for all Lab-PLE scenarios results in a very high heating demand in, for example, heating during polyphenol extraction, and heating during distillation to recover the solvents.

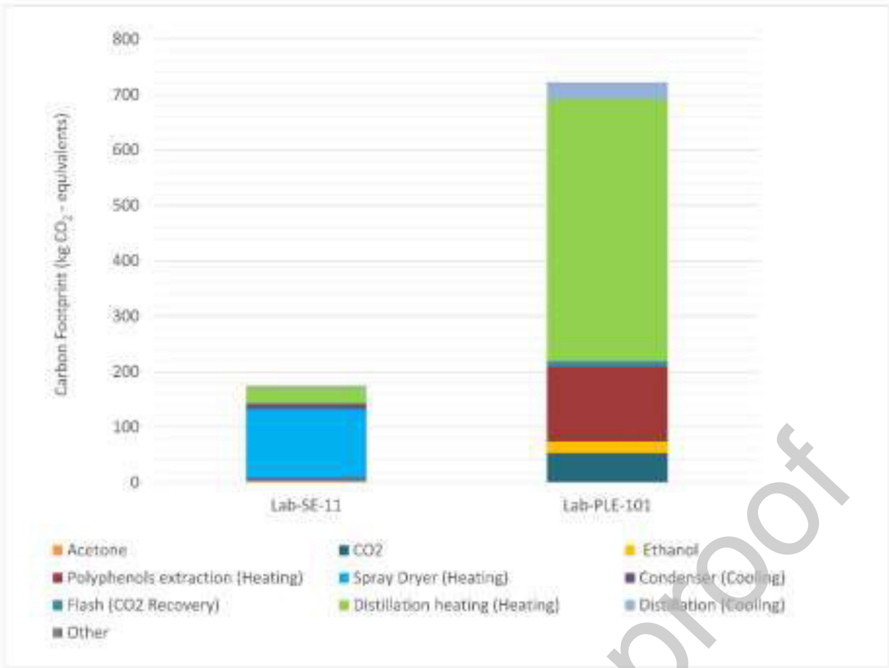

Figure 3 Global warming potential results per kg GAE of polyphenol extraction scenarios at laboratory scale. SE is solvent extraction, while PLE is pressurized liquid extraction. The number at the end of each scenario indicates the solvent to DW ratio for the extraction process.

From the CFP, the importance of keeping the solvent ratio as low as possible is evident. This has a trickledown effect on the energy demand of the whole system. The results can be used in the early design phase, in order to avoid excessive environmental burden later on. By identifying hot spots early on, it is possible to envision adjustments to the production setup, so that the identified hot spots are addressed. Measures, such as increasing the time of contact between solvent and pomace were identified after the CFP. Systems with multiple extraction stages and lower solvent to DW ratios were considered in the TEA.

3.2. Techno-economic assessment of industrial scale processes

The estimated CapEx for the different scenarios are: M€ 6.3 for SE-10, M€ 4.6 for SE-5, M€ 4.5 for SE-2, M€ 25.9 for PLE-50, M€ 16.6 for PLE-25, and M€ 9.8 for PLE-10. For the assessed solvent to DW ratios, the estimated CapEx are significantly higher for PLE compared to SE. Higher solvent ratios require larger equipment and a higher pressure results in more expensive equipment. Due to higher required solvent to DW ratios, the costs related to solvent recovery (i.e. electricity and heat) and solvent supplement are also higher for PLE compared to SE. On the other hand, PLE has a higher extraction yield compared to SE. By looking at processing costs expressed in €/kg GAE (Figure 4), it is clear that the higher extraction yield for PLE does not compensate the higher costs. Only labor related costs are lower for PLE. Scenario SE-2, which has the advantage of a low solvent to DW ratio, has the lowest processing costs. However, because of the required drying step and the low solvent to DW ratio, the assumed extraction yield was considered to be uncertain. As a result, the most feasible options, from a techno-economic perspective, are SE-5 and PLE-10. In the technically feasible range of solvent ratios, SE performs techno-economically better compared to PLE. Details on estimated CapEx, solvent loss, and utility usage for all assessed scenarios is shown in Table S2 of the SI.

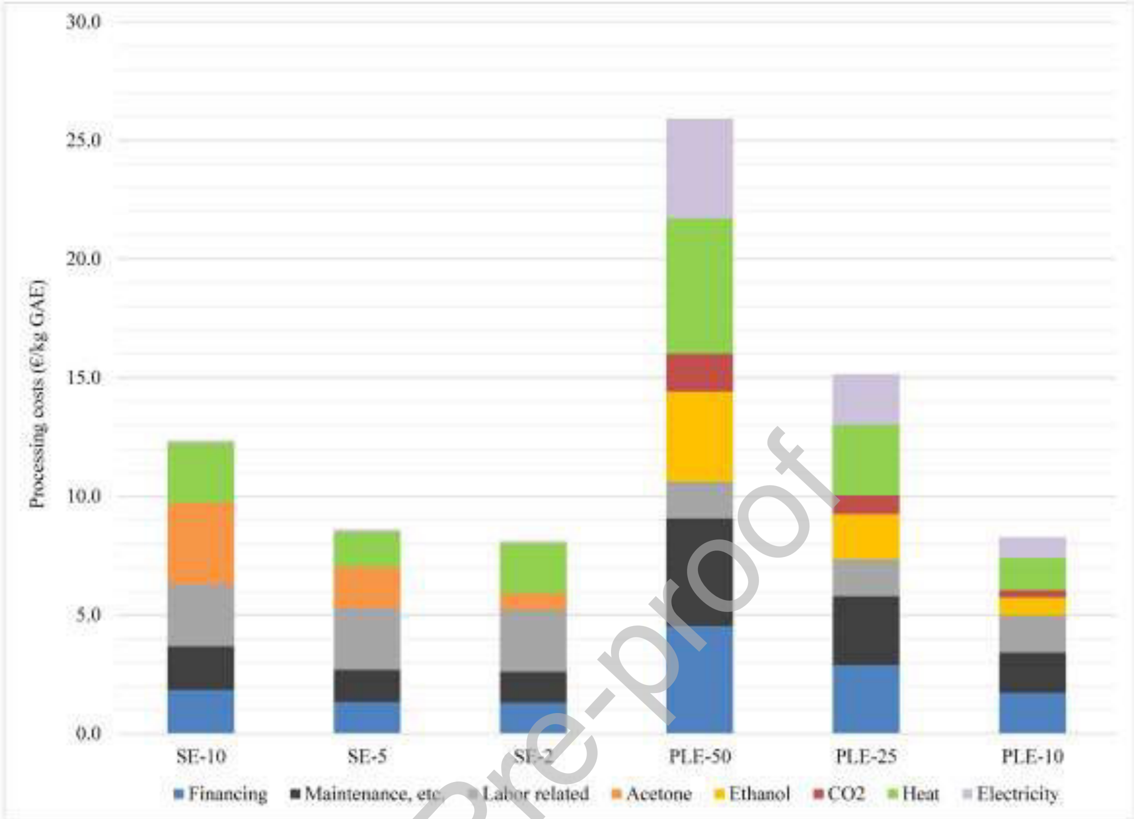

Figure 4 TEA results of polyphenol extraction at industrial scale. SE is solvent extraction, while PLE is pressurized liquid extraction. The number at the end of each scenario indicates the solvent to DW ratio for the extraction process.

3.3. Life cycle assessment of industrial scale design

The LCA of optimized operational conditions showed that if seeking to alleviate GWP it would be preferable to choose SE-2, that is to say, a solvent extraction using acetone with a solvent ratio of 2, Figure 5. However, as mentioned previously, the extraction yield of SE-2 was considered to be uncertain and therefore SE-2 was not considered to be a competitive option. Moreover, PLE-50 and PLE-25 perform far worse than the other options in terms of GWP and all other impact categories (Figure S2, SI), so these are also not deemed competitive options.

From Figure 5 it is possible to see the effect of the optimization performed via process design. The hotspot analysis still points towards solvent quantities as a key parameter for environmental outcomes, e.g. energy used for cooling and heating for distillation dominate the CO_2 burden, and energy for compressing the system. However, through process optimization it is possible to drastically reduce some GWP impacts that were large in the laboratory scale CFP, as for example the impact from the spray dryer for the SE options, by adding a concentration (filtration) step before the drying, which was not part of the laboratory design. On the other hand, it is possible to see that adding a drying step for the pomace in option SE-2, does not pay off in comparison to not drying in SE-5, as the dryer plus distillation heating and cooling, are on the same range of GW impact as just distillation heating and cooling in SE-5. The overall GWP is lower for all options due to the reduction in solvent use and addition of extraction steps.

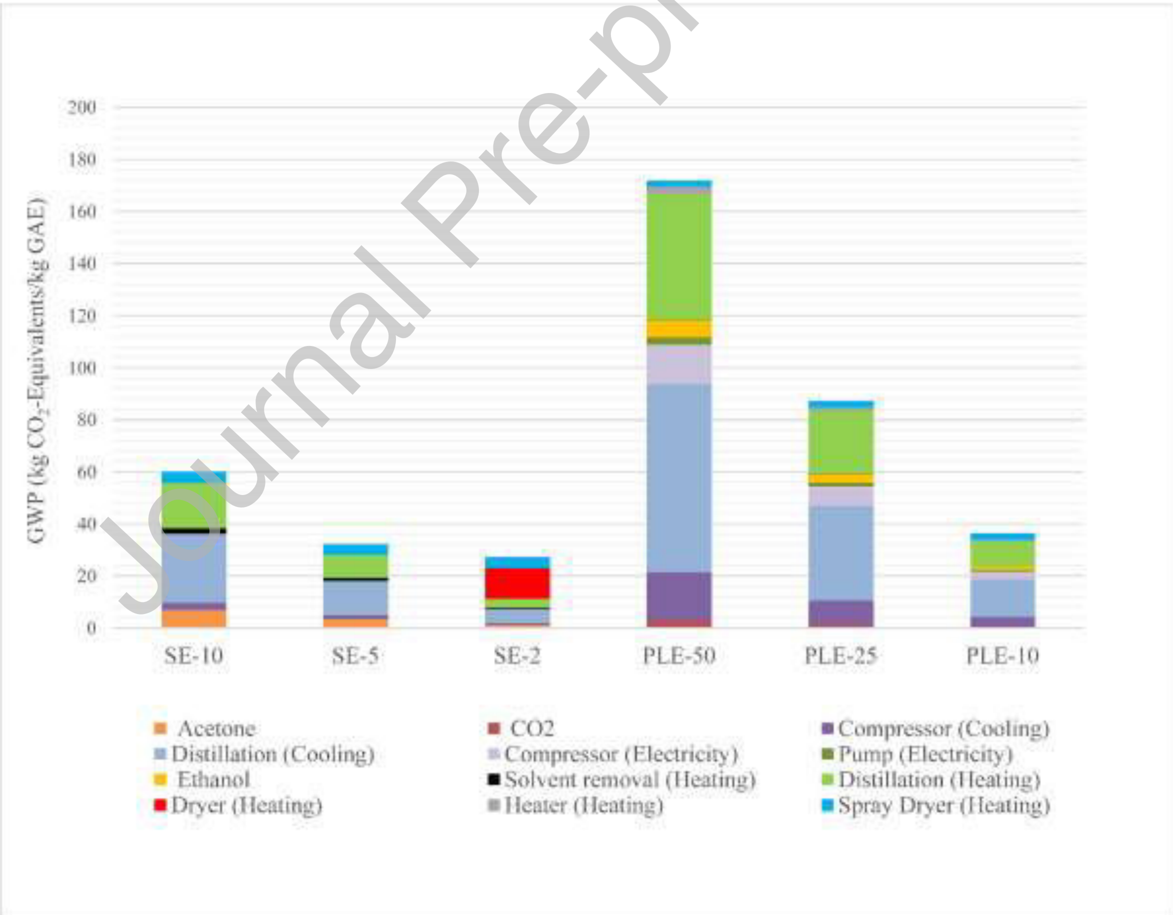

Figure 5 Global warming potential for scenarios tested in kg of CO2-equivalents. Contribution per processing step, cut-

off 1% of overall impact. SE is solvent extraction, while PLE is pressurized liquid extraction. The number at the end of each scenario indicates the solvent to DW ratio for the extraction process.

Results of the TEA show the importance of the solvent to DW ratio for the feasibility of extraction processes. High use of solvent leads to high operational costs and increased demand for electricity and heat, which affect the results of both TEA and LCA. On the other hand, higher yields allow more leeway for higher energy consumption, though not always fully compensating for all GW impacts. A lower solvent to DW ratio results in lower costs for solvent recovery, lower solvent loss, and lower CapEx. These results are mirrored in the LCA, where results benefit from lower solvent use, while midpoint impacts are increased due to the extra heating demand from large solvent volumes. In this regard though, it was clear in the LCA that solvent use, especially if the solvent is acetone, comes with higher GW impacts than electricity or heat use. This is easily illustrated when looking at the CO_2-Equivalents per 1 kg of acetone compared to 1 kg of ethanol or 1 kWh of electricity, as shown in (Table 3). From Table 3 it is possible to visualize that, in terms of the overall LCA assessment, added acetone or ethanol weigh more than added heat or electricity, with acetone being two times more burdensome than ethanol. Nevertheless, the use of solvent in the PLE options is high enough that even though ethanol is less burdensome the total GWP impact outweighs the acetone use in the SE options.

In this regard, it is also worth mentioning that the ethanol used for this assessment is of petrochemical origin. However, since the waste being treated is wine pomace, it is quite possible that a biorefinery treating this waste would also produce bioethanol. This is true for distilleries placed in Italy and France, which currently treat wine pomace in order to produce bioethanol, bioenergy and food additives, among others (Lempereur and Penavayre, 2014). Bio-sourced ethanol will incur different environmental impacts, which were not investigated in this study.

Furthermore, the TEA in this study considers the processing costs including the financing costs. The market price of the product, the extracted polyphenols, and the market volume are yet to be explored. Once a market price or price range is known, then CapEx and processing costs can be compared to the benefits, and profitability indicators, such as net present value and internal rate of return. A larger investment for more complex technology (PLE instead of SE) might be justified if the benefits are significantly larger e.g. a higher yield for PLE than in the present study.

The most competitive options based on all midpoint impacts (Fig S2) and TEA; SE-10, SE-5 and PLE-10, were analyzed further to see if there is burden shifting between environmental indicators and to derive single scores for the options.

3.4. The Single score results

After applying RIF, weighting strings can be derived for the application of TOPSIS with a range of importance given to economic impact from 0-1000, of a sum of 1000 available points distributed in the weighting profile between economic weight and environmental weight (Table S4). The relative importance amongst environmental impacts can also be shown in a single string to improve transparency of the weighting (Table 4).

Table 4 Weighting strings for RIF of environmental impacts used in this study, developed as described in section 2.6.

Impact category	RIF	Impact category	RIF
Fine particulate matter formation	12.14	Marine ecotoxicity	171.22
Fossil resource scarcity	256.66	Marine eutrophication	0.94
Freshwater ecotoxicity	197.95	Mineral resource scarcity	0.004
Freshwater eutrophication	90.31	Ozone formation, Human health	22.35
Global warming	54.5	Ozone formation, Terrestrial ecosystems	26.86
Human carcinogenic toxicity	60.66	Stratospheric ozone depletion	2.06
Human non-carcinogenic toxicity	4.21	Terrestrial acidification	19.86
Ionizing radiation	31.02	Terrestrial ecotoxicity	39.87
Land use	0.6	Water consumption	8.79

This is also done for equal weights (EW) amongst environmental impacts and the same range of importance of economic impact (Table S5). Applying these weightings to the criteria derived from LCA and TEA using TOPSIS, it is possible to provide decision support in the form of a single score indicator of idealness of the various technological alternatives including a relationship to environmental relevance across all impacts (Figure 6). Furthermore, based on the results of the application of TOPSIS, a preference ranking can be made, with PLE-25 ranked fourth, SE-10 ranked third, and either PLE-10 or SE-5 ranked first and second. The ranking for first and second is based on the weight given to economics in the decision making process.

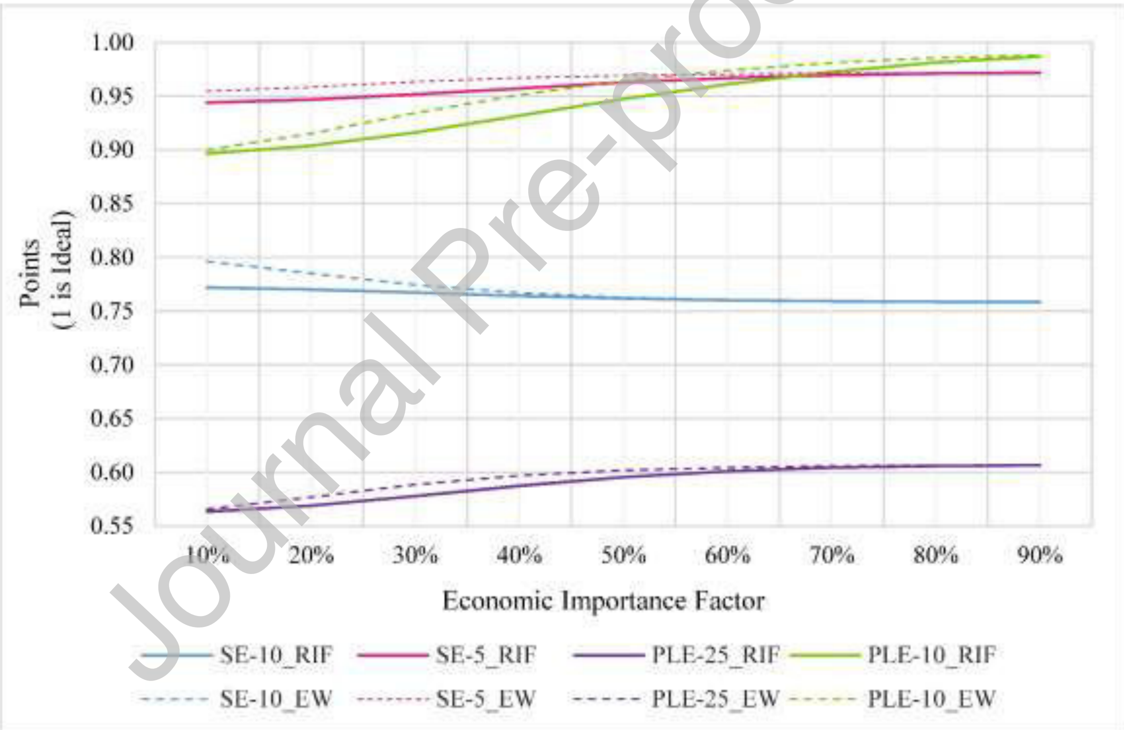

Figure 6 TOPSIS derived single score indicator of idealness (most ideal=1) for both Relative Importance Factor (RIF) derived environmental weighting and Equal Weights (EW) environmental weighting amongst a range of weights given to economic performance. SE is solvent extraction, while PLE is pressurized liquid extraction. The number in each scenario indicates the solvent to DW ratio for the extraction process.

Based on the application of TOPSIS, it can be easily concluded that the PLE-10 and SE-5 methods outperform all other alternative extraction methods. While PLE-10 is the best economic performer, SE-5 proved to be the best environmental performer, though the differentiation between these is likely below the potential margin for error. This results in a shift in decision support depending on the weight given to economic factors, but again, this differentiation is likely not statistically significant. In addition, SE-10 consistently performs better than PLE-25 both environmentally and economically. This differentiation is statistically significant across all ranges of economic weighting. This results in a preference of SE-10 over PLE-25 regardless of weight given to economics. And, given that it is likely that an industrial process would be developed with a solvent ratio between the minimum and maximum solvent ratios as shown here for each technology respectively, it is apparent that there is more likelihood for SE to outperform PLE across all economic weightings (see SI figure S3).

As can be seen in Table 4 and Table 5, there is significant range in the importance of specific environmental impacts in RIF for the assessed methods. For example, some impacts such as human non-carcinogenic toxicity, marine eutrophication, and land use are insignificant in relative importance, and mineral resource scarcity is almost entirely irrelevant. On the other hand, fossil resource scarcity and freshwater ecotoxicity make up nearly half of weighting applied to environmental impacts due to the scale of their impact compared to the other environmental criteria relative to the average European's environmental impact.

One other element of note is the difference of decision support between EW and RIF in terms of the importance given to economic impact when PLE-10 is preferred over SE-5. When applying the RIF, this switch in preference occurs at appx. 65% weight to economic factors while for EW, the switch occurs at 55%. This is primarily due to the effective removal of environmental impact categories where the two alternatives are relatively equal that were compensating for other impact categories where the technologies were less equal in terms of performance. This occurs through the application of the ArgCW-LCA RIF weighting (Table 5) because some impact categories do not present much relevance to the decision context. This can be because there is either very little variation of the particular impact category amongst the assessed alternatives or because the given impact is smaller relative to status quo per capita emissions in relation to the other impacts of the assessed system.

Table 5 Relative weight (RW) of environmental impacts between RIF and EW weighting ($RW = W_{RIF}/W_{EW}$)

Impact category	RW	Impact category	RW
Fine particulate matter formation	21.85%	Marine ecotoxicity	308.19%
Fossil resource scarcity	461.99%	Marine eutrophication	1.69%
Freshwater ecotoxicity	356.31%	Mineral resource scarcity	0.01%
Freshwater eutrophication	162.56%	Ozone formation, Human health	40.23%
Global warming	98.10%	Ozone formation, Terrestrial ecosystems	48.35%
Human carcinogenic toxicity	109.18%	Stratospheric ozone depletion	3.70%
Human non-carcinogenic toxicity	7.57%	Terrestrial acidification	35.76%
Ionizing radiation	55.84%	Terrestrial ecotoxicity	71.77%
Land use	1.07%	Water consumption	15.83%

Another important element in interpreting the results from RIF weighting is understanding that there is a level of uncertainty in the normalization factors used to derive the RIF, and that the decision to use current emissions as a reference point, i.e. by using a European's environmental impact as NF, does not necessarily have a relationship to the severity or consequences of environmental impacts. However, it does provide an indication of the relative importance of an emission, or reduction thereof, to the status quo. If absolute sustainability related factors were available for all relevant impact categories, the application of these instead of normalization factors would be preferable, as they would provide a stronger link to environmental impact. Ideally, this process would be completed relative to planetary boundaries (Steffen *et al.*, 2015) using an absolute relationship to impacts from LCA (Bjørn *et al.*, 2015). However, this cannot be done because this absolute relationship is not yet well enough understood/developed, nor has it been developed to include all impact categories covered in LCA.

An alternative to either of these methods would be to derive a RIF weighting from endpoints using e.g. monetization. While this might seem appealing, as there is a stronger connection with environmental damages when using endpoint indicators in LCA, the challenge comes in determining the relative importance of the different damage categories. This relative importance is purely subjective, and as such a specific cultural perspective would be applied to the derivation of the weighting profile. While this could be carried out in a scientific fashion to be representative of a decision maker group, the results would already contain some bias toward certain impacts introduced in the endpoint calculation (Kalbar et al., 2016; Sohn et al., 2017). This would make the results more challenging to interpret and potentially lead to decision support that in the end does not reflect the true preferences of the decision maker. And, though midpoint impacts are not devoid of subjectivity, utilizing RIFs based on midpoint impacts effectively reduces the layers of interpretation applied in the interpretation phase of the impact assessment relative to endpoint derived single scores. Thus, making the elements driving decision support easier to track and understand.

4. Conclusions

Polyphenol extraction methods were assessed using LCA at laboratory scale and a combination of TEA and LCA for designed industrial scale processes. Solvent to DW ratio and extraction yield are important parameters considering the design of the industrial scale processes, and therefore have a large impact on the results of the TEA and LCA. Thus, it is recommended that these parameters are optimized in the laboratory to ease their translation into industrial scale processes.

Out of the solvent to DW ratio ranges of the TEA-LCA, SE options have potential to perform better than PLE. Despite higher yields for PLE, higher economic and environmental burdens outweigh the benefit of higher yield for this option. The most important parameters indicated by the TEA are the polyphenol extraction yield and the solvent to DW ratio. The most important parameter for optimization indicated by the LCA results is reducing solvent amounts. The CFP at laboratory scale was useful in pointing out potential environmental hotspots, which served to guide the design of the industrial scale processes, from both an economic and environmental perspective. The single score indicator concluded that the potential performance is better when utilizing SE-5 than PLE-10, though a shift in preference is seen for higher economic weight. The addition of a transparent and reproducible decision assessment process aided in the understanding of the holistic impacts of the alternatives And, it can be concluded that the introduction of RIF as a method of deriving a weighting, relative to equal weights, for use in MCDA for LCA can likely reduce the impact of irrelevant and/or subjective criteria on the conclusions drawn from the application of MCDA that include weighting such as TOPSIS. Furthermore, based on the application of TOPSIS, assuming that PLE-25 and SE-10 represent presently attainable solvent to DW ratios, while PLE-10 and SE-5 represent future potentially attainable solvent to DW ratios, it can be concluded that there is greater potential for better performance utilizing solvent extraction than pressurized liquid extraction across all value scales relating the environment and economics.

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Declaration of interests

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

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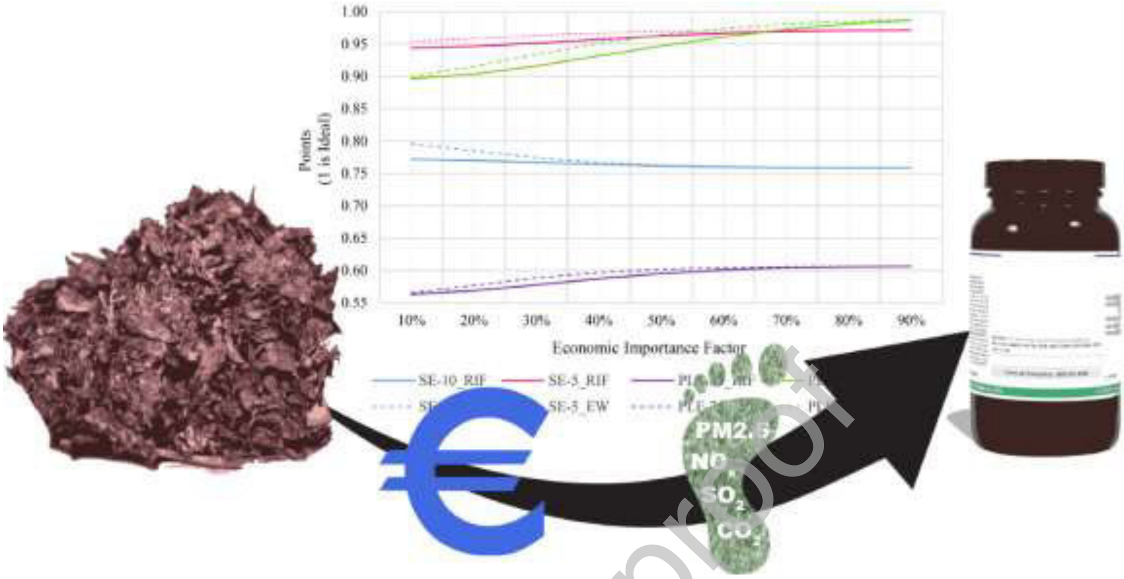
Graphical abstract



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SUSTAINABILITY OF FOOD P&P SYSTEMS

Using life cycle assessment to quantify the environmental benefit of upcycling vine shoots as fillers in biocomposite packaging materials

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Abstract

Purpose The objective of the present study was to better understand the potential environmental benefit of using vine shoots (ViShs), an agricultural residue, as filler in composite materials. For that purpose, a comparative life cycle assessment (LCA) of a rigid tray made of virgin poly(3-hydroxybutyrate-co-3-hydroxyvalerate) PHBV, polylactic acid (PLA) or polypropylene (PP), and increasing content of ViSh particles was performed. The contribution of each processing step in the life cycle on the different environmental impacts was identified and discussed. Furthermore, the balance between the environmental and the economic benefits of composite trays was discussed.

Methods This work presents a cradle-to-grave LCA of composite rigid trays. Once collected in vineyards, ViShs were dried and ground using dry fractionation processes, then mixed with a polymer matrix by melt extrusion to produce compounds that were finally injected to obtain rigid trays for food packaging. The density of each component was taken into account in order to compare trays with the same volume. The maximum filler content was set to 30 vol% according to recommendations from literature and industrial data. The ReCiPe 2016 Midpoint Hierarchist (H) methodology was used for the assessment using the cutoff system model.

Results and discussion This study showed that bioplastics are currently less eco-friendly than PP. This is in part due to the fact that LCA does not account for, in existing tools, effects of microplastic accumulation and that bioplastic technologies are still under development with low tonnage. This study also demonstrated the environmental interest of the development of biocomposites by the incorporation of ViSh particles. The minimal filler content of interest depended on the matrices and the impact categories. Concerning global warming, composite trays had less impact than virgin plastic trays from 5 vol% for PHBV or PLA and from 20 vol% for PP. Concerning PHBV, the only biodegradable polymer in natural conditions in this study, the price and the impact on global warming are reduced by 25% and 20% respectively when 30 vol% of ViSh are added.

Conclusion The benefit of using vine shoots in composite materials from an environmental and economical point of view was demonstrated. As a recommendation, the polymer production step, which constitutes the most important impact, should be optimized and the maximum filler content in composite materials should be increased.

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1 Introduction

In viticulture, every winter after pruning, large quantities of vine wood are produced that are currently underutilized. Pruning of vine shoots (ViShs) is necessary in order to improve growing conditions for the plant, as well as to increase the yield and quality of grapes. Vine shoots can be from 1 to 2 m long, and production amounts to between 1 and 2.5 t of dry matter per hectare per year (Galanakis 2017). The productivity of the vine plant depends on the region where it grows, the pruning method, and the vine species. In Languedoc-Roussillon (LR), a wine region in the south of France, ViSh production amounts to 500,000 t per year (IFN, FCBA, Solagro 2009). Currently, management of vine shoots in France is done by either collecting and burning the ViSh or leaving them on the vineyards where they are rough-cut and used as organic fertilizer (FranceAgriMer 2016). When used as biofertilizers, ViSh should be considered by-products and not waste. However, their use as soil amendment can be problematic, as decomposing ViSh may serve as vector for diseases for the following vine crop (Chambre régionale d'agriculture Nouvelle-Aquitaine and DRAAF/SRAL Nouvelle-Aquitaine 2017). Furthermore, it is worth noting that ViSh is not the most judicious biofertilizer since its biodegradation, i.e., mineralization in soil, competes with the vine's growth with regard to nitrogen consumption (Keller 2015). Less commonly, ViShs are used as fuel wood or compost, which are considered low-value uses for this potential resource. Regarding the ambitious goals set by the European community for a bioeconomy, which include the decarbonization of the economy through an 80-95% decrease of CO_2 emissions by 2050 (Scarlat et al. 2015), ViShs present a valuable resource for implementing decarbonizing recovery strategies. These strategies can be achieved in a biorefinery context, where cascading treatments of ViSh are investigated to produce value-added products, including the production of lignocellulosic fillers for biocomposite applications (Kilinc et al. 2016; David et al. 2019, 2020a). Lignocellulosic fillers from agricultural residues present the advantages that, in addition to their fully biodegradability in natural conditions, they have a lower density than conventional inorganic fillers and are highly available at a low price, with no competition from the food sector (Mohanty et al. 2001). ViShs present a great opportunity in the field of biocomposites, with a potential application being rigid food packaging that is biodegradable in natural conditions (David et al. 2020c; Guillard et al. 2018).

On the other hand, the global plastic market is continuously growing having reached 350 million tons in 2018, with 40% of the production used in the packaging sector (PlasticsEurope 2018). The massive amount of plastics used each year results in a constant accumulation of plastic wastes in our environment (Geyer et al. 2017). The associated effect of this on ecosystems, wildlife, and humans is worrying, if not yet fully understood. For this reason and the concern about global warming, fully bio-sourced and biodegradable materials such as biocomposites are emerging as a possible solution to tackle the problem of accumulation of plastic in our environment and to reduce greenhouse gas emissions. Poly(3-hydroxybutyrate-co-3-hydroxyvalerate), called PHBV, is a promising bacterial biopolymer that is biodegradable in the soil and ocean, and that can be synthetized from many types of carbon residues. PHBV can be combined with natural fillers to create fully biodegradable biocomposites, e.g., for application in rigid trays (Berthet et al. 2015a; Lammi et al. 2018). Moreover, PHBV displays similar mechanical and barrier properties as polyprolylene (PP) and can therefore act as a viable substitute for this fossil-derived and non-biodegradable conventional polymer (Chodak 2008). A competitor to PHBV is polylactic acid (PLA), which is the most widely commercialized bio-sourced plastic currently in the market. However, it is worth noting that PLA is not fully biodegradable in natural conditions, but only compostable in industrial conditions (Gurunathan et al. 2015), which requires collection and sorting in order to achieve a valuable end of life management and does not avoid concerns related to plastic accumulation from littering or leakage.

The development of biocomposites is largely motivated by either an improvement of the overall technical performance, the need for specific mechanical properties, a decrease of the overall cost of materials, and the improvement of the carbon footprint, by replacing a part of non-renewable fossil resources (Mohanty et al. 2005). Biocomposites are thus generally presented as eco-friendly materials. However, most of the time, the environmental benefit is not quantitatively proven (Civancik-Uslu et al. 2018). It is thus necessary to ensure that the biocomposites are actually capable of mitigating the abovementioned environmental problems, as the use of bioplastics and natural fillers to produce biocomposites does not automatically make them sustainable. In order to quantitatively verify environmental claims made about biocomposites and other innovative materials, it is possible to carry out environmental assessments.

Life cycle assessment (LCA), which is a holistic tool capable of measuring environmental impacts of products and services, can be applied to emerging biomaterials (Hauschild et al. 2018). It investigates the inputs (i.e., resources and energy) and outputs (i.e., waste gases, wastewater, and solid waste) across the entire life cycle stages (cradle-to-grave).

LCA allows location of "hot spots" in the life cycle and avoids the shifting burdens from one life cycle stage to another while accounting for all types of emissions and resource consumption (Qiang et al. 2014). Its main limits are the collection of data, which can be difficult, and the initial assumptions that need to be justified. Most of the LCAs carried out for biocomposites focus on the comparison of natural fillers with synthetic fibers (Kim et al. 2008; Le Duigou et al. 2011; Civancik-Uslu et al. 2018), especially for applications in the automotive industry (Joshi et al. 2004; Duflou et al. 2012; Boland 2014). Generally, natural fillers tend to have a better environmental performance than glass fibers, notably thanks to the weight reduction of the composites and their low energy demand for production (Joshi et al. 2004).

There are fewer papers in the literature regarding the environmental advantage of incorporating natural fillers in polymer matrices. In a previous study considering 1 kg of material as functional unit, the environmental impacts of materials made of virgin polyolefins (PP and HDPE) and biocomposites with natural fillers (derived from rice husks and cotton linters) were compared (Vidal et al. 2009). The LCA showed that composites displayed lower environmental impacts in all impact categories, except eutrophication, due to the use of fertilizers for rice cultivation. Similarly, it was shown that the incorporation of either wood flour or wood fiber allowed for reducing the environmental impacts of HDPE (Xu et al. 2008) and PP (Xu et al. 2008), respectively, in proportion to the filler content.

LCAs of vine shoots and their incorporation in composites were not found in the literature. The combustion of ViSh and induced emissions have previously been studied (Spinelli et al. 2012; Picchi et al. 2013) without LCA tools. More recently, Gullón et al. performed a LCA of the valorization of vine shoots into antioxidant extracts, and other bioproducts from a biorefinery perspective (Gullón et al. 2018). They determined that ViSh production-related processes should be burden-free in the biorefinery system since the environmental impacts were entirely allocated to the grape harvesting, as ViShs were considered agricultural waste (Sanchez et al. 2002; Max et al. 2010).

Concerning PHBV, no process data is currently available in the Ecoinvent database. However, as shown by Yates and Barlow (2013), several LCAs about bioplastics including PHBV are available in the literature. Inventory data from these papers can be used (Harding et al. 2007; Yates and Barlow 2013).

In this context, the objective of the present study was to better understand the potential environmental benefit of using vine shoots as raw resources for the production of lignocellulosic fillers for biocomposite applications. For this purpose, a comparative life cycle assessment was carried out, first on rigid trays made out of virgin PHBV, polylactic acid (PLA), or polypropylene (PP). Then, the effect of ViSh incorporation in these 3 polymer matrices was studied, utilizing a cradle-to-grave approach. The contribution of each life cycle step was identified and discussed. Furthermore, the balance between the environmental and the economic benefits of composite trays was discussed.

2 Methodology

2.1 Goal and scope

The aim of this article was to determine to what extent addition of ViSh fillers in packaging trays was environmentally beneficial when compared with trays produced entirely from virgin plastics. For that purpose, the environmental performance of packaging trays produced in France from either 100% virgin plastics or related ViSh-based biocomposites was assessed. Composites with three polymer matrices, i.e., PHBV, PLA, and PP, and different filler contents were compared. The ReCiPe 2016 Midpoint Hierarchist (H) method was used during the impact assessment phase. All background data used in the assessment were obtained from the Ecoinvent v.3.4 database (Wernet et al. 2016) with the cutoff system model and processed using the LCA software Simapro v.8.5 (PRé Sustainability 2018). The cutoff approach was chosen to reduce the potential for conflating information and to simplify the product system. Based on the "cutoff" approach, the used product from a first life is considered to be waste that does not bear any environmental burden from previous life.

The functional unit was a tray of standard model (176 × 162 × 40 mm, GN 1/6 type), 25 cm^3 in volume, for single-use packaging, produced by injection molding. It was assumed that all the considered trays had the sufficient properties to provide the same service. The volume of the trays was thereby kept equal throughout the assessment. However, due to the intrinsic densities of the considered materials, the final weight of the trays varied according to the nature and the proportion of each constituent. The scenarios included in this study were trays of virgin PHBV, PLA, and PP, and trays of PHBV, PLA, and PP filled with milled vine shoots.

Figure 1 displays the system boundary considered in the present study, with the different life cycle steps that were included. It was assumed that the collection of vine shoots and the production of the trays were done in the Languedoc-Roussillon region of France. In the case of 100% virgin plastic trays, the steps encased by dashed lines in Fig. 1 were irrelevant because they concerned the ViSh treatment and compounding steps.

The pruning is a necessary process that is independent from the fate of the ViSh. It is difficult to estimate the exact proportion of burnt ViSh because this practice, which is a common fate for ViSh, is in theory forbidden, but derogations and tolerances still exist (Ministère de l'écologie et du

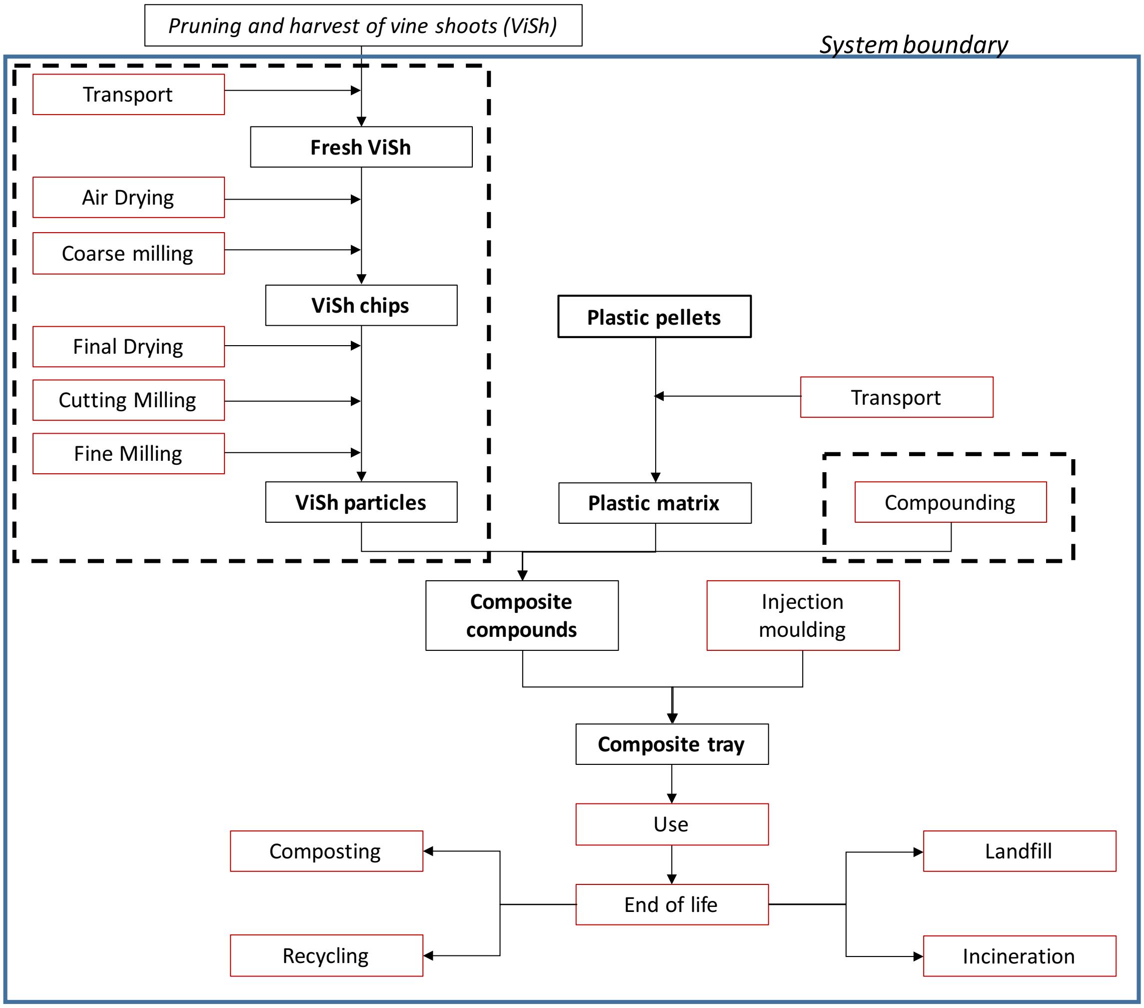


Fig. 1 Boundary of the studied system

développement durable 2011). According to FranceAgriMer, burning of ViSh accounts for between 25 and 50% in France (FranceAgriMer 2016; Gazeau et al. 2018). In the present study, ViShs burnt on site or without valorization were considered. In that case, the collection of the ViSh happens anyway in order to remove ViSh from the vineyards and it was therefore considered a part of the grape cultivation production system. Furthermore, ViShs have no market value, and thereby, zero environmental impact would be allocated to them if allocation were to be used. ViShs were, thus, considered burden-free in the present system. Additionally, ViSh being produced in a wine-grape production system, all the environmental impacts of production were ascribed to the production of wine grapes. Therefore, pruning and harvest of ViSh were considered out of the system boundary.

The main properties of the raw materials are presented in Table 1. They correspond to commercial-grade PHBV (PHI002 from Natureplast), PLA (PLI 003 from Natureplast), and PP (PPH9020 from Total Petrochemical). The density of ViSh was experimentally determined, as explained in Supplementary Data.

It was previously shown that increasing the content of ViSh in PP (Girones et al. 2017), PE (Girones et al. 2017), or PHBV (David et al. 2020b) resulted in a slight decrease of the mechanical properties of the materials. Ahankari et al. (2011) studied the reinforcement of PHBV and PP with agro-residues and recommended to incorporate filler contents lower than 40 wt% to avoid a decrease in mechanical properties, due to an increased filler agglomeration in the polymer matrix. Confirming this, Berthet et al. (2015b) observed that the processability of PHBV/wheat straw biocomposites became difficult when the filler content was above 40 wt%. Authors usually considered weight filler contents. However, considering that the volume of the injected molding tray remains constant whatever the matter, it was considered that the use of volume filler contents was more pertinent to compare the different formulations. Given that, it was assumed that the maximum ViSh filler content to attain satisfactory physical

Table 1 Different properties for the components of the studied biocomposites.

	Density ($g\ cm^{-3}$)	Weight (g) (25 cm^3 tray)	Melting temperature (°C)	Degradation (°C)	Young's modulus* (GPa)	Stress at break* (%)	Strain at break* (%)
PHBV	1.23	30.75	170	200	4.2	40	3.2
PLA	1.24	31	150	250	3.5	45	3
PP	0.91	22.75	165	320	1.7	37	8
ViSh	1.36	-	-	230	na	na	na

*According to the standard ISO 527

properties for the tray application was 30 vol% for all the composites. This was also in accordance with the filler content currently used in commercialized composites (*Vitis Valorem*, Meursault, France, PLA or PP-Sarmine® products). This set limit of 30 vol% corresponded to a weight content of 32 wt% for PHBV and PLA, and 39 wt% for PP (for a given filler volume content and a tray volume, the filler weight content depends on the density of each constituent).

2.2 Life cycle inventory

The inventory is based on figures derived from communications with different industrial producers: one company expert in micronization of powder (*SD-Tech Group*, Alès (Gateau 2019)); one company specialized in the valorization of vine shoots into biocomposites (*Vitis Valorem*, Meursault (Grangeot 2019)); the French technical center of plastics and composites (CT-IPC, Bellignat (CT-IPC 2019)); and one company expert in the injection of plastic trays (Fürstplast, Fourques (Hreblay 2019)). They were interviewed from January to June 2019. The data that was collected was analyzed, compared with theoretical figures, and then finally selected. After this collection of data, existing processes from the Ecoinvent database were adapted to fit the collected data. In accordance with the geographical boundary of the assessment, all the electricity used in the foreground systems was assumed to be provided by the French energy mix.

2.2.1 Raw materials

Polymer matrices were PHBV, PLA, and PP. Ecoinvent processes data recorded for fossil-based PP and PLA from maize grain were used in the LCA. Inventory for PHBV made from sugar cane was obtained from the work of Harding et al (Harding et al. 2007). Transport of plastic matter to the production facility was taken into account using the "Background data for transport" sheet from Ecoinvent as the specific transport mode was unknown (Borken-Kleefeld and Weidema 2013).

For all tested scenarios, lignocellulosic fillers were obtained from the dry milling of ViSh collected in the Languedoc-Roussillon region. In keeping with status quo practices, ViShs were collected during the winter after pruning and initially had a moisture content of 40 wt% (w.b.).

Transport of ViSh from the field to the filler producing site was assumed to be done by a 3.5–7-t lorry with an average distance of 10 km according to *Vitis Valorem* (France) information (Grangeot 2019).

2.2.2 Production of biocomposite trays

Practical information about the handling of ViSh as raw material for the production of biocomposites was provided by *Vitis Valorem* (France) (Grangeot 2019). Commonly, ViShs are first air-dried outdoors for 7 months, between January and August. The corresponding land use was determined considering that the ViShs are arranged on the ground reaching an average height of 2 m, with an apparent density of 30 kg m^{-3}. Only manual labor was used during this step. At the end of this period, the moisture content of ViSh was 20 wt% (w.b.).

Coarse milling with a common wood chipper (Greentec 952, Ufkes Greentec BV, Netherlands) was utilized to mill the ViSh. The throughput was set at 2000 kg h^{-1}, and 10% of the initial ViSh mass were assumed lost during the milling process. Output chip sizes ranged between 3 and 6 cm in their largest dimension. The output is called "ViSh chips."

An additional drying step was required to reduce the moisture content of the ViSh to 5 wt% (w.b.) after air drying. An existing drying process from the Ecoinvent database was used (see Supplementary Data), modified to utilize the French electricity grid.

After coarse milling, a finer milling process in two steps is needed in order to obtain particles of between 0.3 and 0.05 mm in size. First, ViShs were milled using a cutting mill type SM 300 (Retsch, Germany) with a 2.0-mm sieve, and secondly, they were milled with a fine impact mill (CUM 150, Netzsch Condux, Germany). The final output is hereafter called "ViSh particles." Data for milling were provided by *SD-Tech Group* (Alès, France) (Gateau 2019).

Flexible intermediate bulk containers (FIBC, commonly known as "Big Bags") were used to store the ViSh chips after coarse milling, ViSh particles after fine milling and composite granules after compounding. It was assumed that each FIBC was used 3 times per year during a period of 5 years before being discarded. Each FIBC had a mass of 2.5 kg with a capacity of 1 m^3, and it is made from PP. ViSh chips after coarse milling, finely milled ViSh particles and composite granules had a bulk apparent density of 200 kg m^{-3}, 420 g m^{-3}, and 700 g m^{-3}, respectively.

During the compounding step, the plastic was mixed with ViSh fillers in an extruder. The extrusion process in Ecoinvent was adapted with data from *Vitis Valorem* (Grangeot 2019), which uses a compounder, model ZSE 160 HP (Leistritz, Nuremberg, Germany). Electricity consumption of the compounding step was 300 kWh $t^{-1,}$ and the yield is 97.6%. In the assessment, the same yield and energy data is used for all compounding regardless of composite granule type. No plasticizers nor additives were used.

It was assumed that the trays were produced by injection molding of compounds. The injection molding process in Ecoinvent was modified to incorporate provision of electricity from the French electricity mix. The yield was assumed to be 99.4% because scrap and waste could be recycled in a nearly closed loop.

It was further assumed that all of the previously described steps (from air drying to injection molding) occurred at the same location. Table 2 recaps the data collected and used in the inventory of the production of biocomposite trays.

2.2.3 Use phase

It was assumed that the use phase of the biocomposite trays was comprised of the transport from the factory gate to the place at which they are used as food packaging and then to the distribution site. This transportation was assumed to be done utilizing a 32-t lorry with an average distance of 100 km for each transport stage (Labouze and Le Guern 2007). The use by the consumer was assumed to be the same for all assessed materials and thus was omitted from the assessment.

2.2.4 End of life

The end of life (EoL) of each tray was defined according to French practices for municipal waste (ADEME 2018) and considering the characteristics of the materials and existing facilities (Table 3). With regard to transport in the end of life, it was estimated that the trays traveled on average 100 km from household to a waste treatment center (Beigbeder et al. 2019). Transport was assumed to happen by a 16–32-t lorry, EURO5 from Ecoinvent.

Concerning composting, only industrial compositing was included due to the lack of data for home composting. The incineration process from Ecoinvent was adapted to account for CO_2 emissions and the origin of carbon (biogenic or fossil). Anaerobic digestion could be an end of life option for bioplastics and biocomposite trays, but was not included in the possibilities because it is not widely used in France, and it is more dedicated to agricultural wastes than composite materials.

A more detailed inventory for the production of biocomposites is given in the supplementary inventory (SI) of this paper.

3 Results and discussion

3.1 Environmental impact of 100% virgin plastic trays: comparison of PHBV, PLA, and PP

First, the environmental performance of 100% plastic trays without ViSh fillers was compared (Fig. 2). Trays made of

Table 2 Foreground data collected concerning the production biocomposite trays

Step	Foreground data collected	Comments	Unit of the process	Source
Air drying	Duration: 7 months Height of the pile: 2 m Density of ViSh: 30 kg/m^3	Moisture content from 40 to 20 wt%	m^2a (square-meter-years, land use occupation)	Vitis Valorem
Coarse milling	Throughput: 2000 kg/h Yield: 90%	Ref: Greentec 952	h (duration)	Vitis Valorem
Drying step	Yield: 100%	Moisture content from 20 to 5 wt%	l (volume of evaporated water)	Vitis Valorem
Cutting milling	Throughput: 30 kg/h Yield: 99% Nominal power machine: 3 kW	Ref: SM 300 Retsch	kg (mass of matter to transform)	SD-Tech
Fine milling	Throughput: 29 kg/h Yield: 99% Nominal power machine: 7.5 kW	Ref: CUM 150 Netzch Condux	kg (mass of matter to transform)	SD-Tech
Compounding	Yield: 97.6% Electricity consumption: 300 kWh/t	Ref: ZSE 160 HP Leistritz	kg (mass of matter to transform)	Vitis Valorem
Injection molding	Ecoinvent data “injection molding {RER}| processing”		kg (mass of matter to transform)	EcoInvent

Table 3 Current possible end of life of the different trays (in weight %) from (ADEME 2018)

Tray material	Landfill (%)	Incineration (%)	Recycling (%)	Composting (%)
PP	34.6	36.5	28.9	0.0
PP-ViSh composite	48.7	51.3	0.0	0.0
PHBV	38.0	40.0	0.0	22.0
PHBV-ViSh composite	38.0	40.0	0.0	22.0
PLA	38.0	40.0	0.0	22.0
PLA-ViSh composite	38.0	40.0	0.0	22.0

PP displayed lower impacts than those of PLA or PHBV trays in all the midpoint impact categories except for *fossil resource scarcity*. This could be explained by the fact that the density of PP (0.91 g cm^{-3}) was lower than those of PHBV or PLA (1.23 and 1.24 g cm^{-3}, respectively). Thus, in order to get the same tray, i.e., with the same volume, a smaller amount of PP (in mass terms) was needed, i.e., 22.75 g instead of 30.75 g for PHBV (Table 1). Similar results were found showing that when compared by volume rather than weight, PHBV had higher environmental impacts than PP or PE (Tabone et al. 2010). Moreover, the production of 1 kg of PHBV or PLA induced higher impacts than the production of PP. Impacts for *stratospheric ozone depletion*, *freshwater and marine eutrophication*, *land use*, and *water consumption* were very low for PP, in comparison with the assessed bioplastics. This is primarily because the life cycle of PP does not have agriculture activities, which, in this assessment, heavily contributed to the above named impact categories. On the other hand, the *fossil resource scarcity* impact for PP was the highest, at least in part, because PP is entirely made from fossil resources. In regard to the assessed bioplastics, results showed that PLA induced the greatest impact for 13 out of the 18 impact categories.

The impact of each production step on *global warming* is presented in Fig. 3. The production of polymer pellets was the largest contributor to induced impacts throughout the life cycle of a plastic tray, accounting for more than half of the burden for PP and more than 80% for PLA and PHBV. The PP tray impacts were 30% lower compared with bioplastic trays. This suggests that the substitution of traditional plastic trays with bio-based materials does not always result in a lower environmental impact. Nevertheless, conventional plastic industries have a high degree of optimization, which is not the case for bioplastics that are produced in low tonnage with relatively less developed technologies. This is exemplified by PP, a petrochemical matrix polymer, for which the production has been highly improved over nearly 70 years of development, whereas the development of biopolymers is recent; thus,

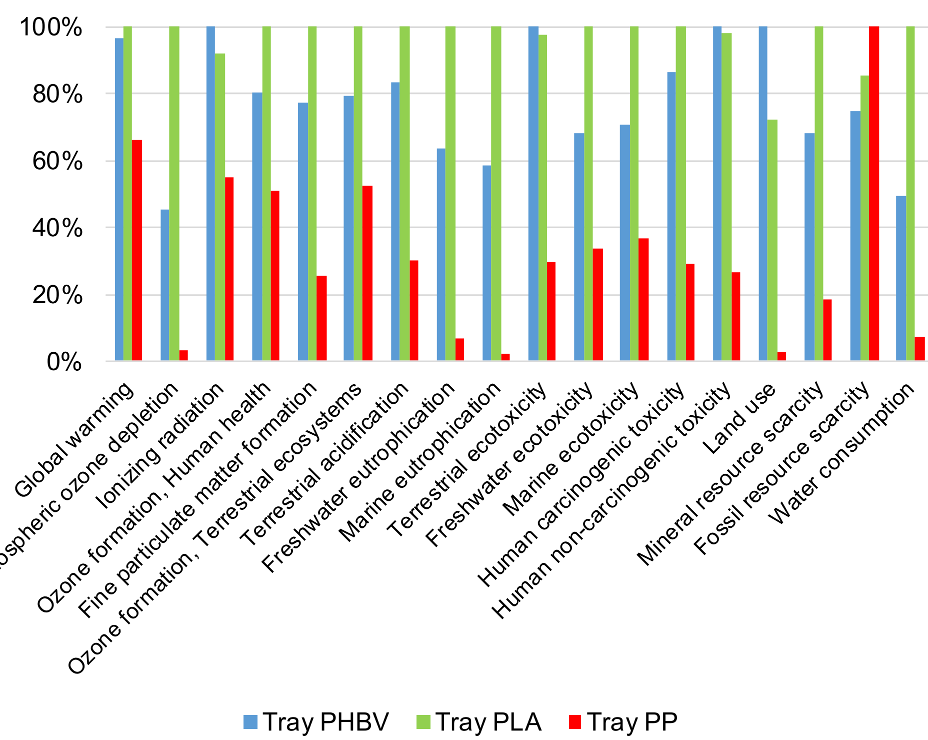


Fig. 2 Environmental impact for all impact categories of the ReCiPe 2016 (H) method, for 100% virgin plastic trays

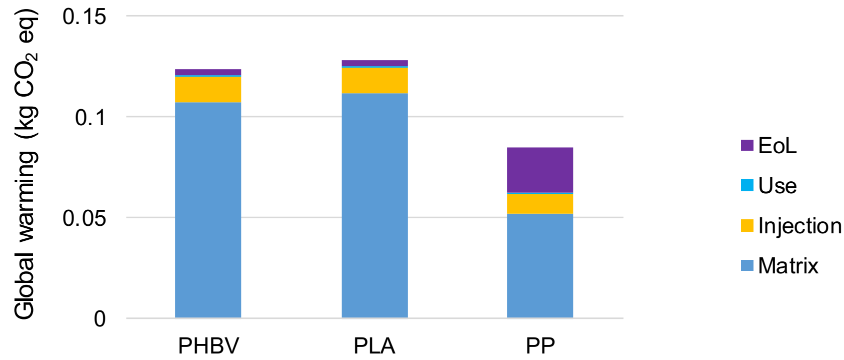


Fig. 3 Global warming impact of one 100% plastic tray (without fillers)

they have not yet reached the same level of technological maturity. This leads one to the determination that further research on the optimization of the bioplastics processing toward their environmental improvement should be conducted (Vidal et al. 2009). Therefore, it is expected that the environmental impacts induced by the production of bioplastics will be smaller than those observed in status quo production—and thus less than the impacts exhibited by the production reflected in the present study.

The use phase was not a large contributor to the overall life cycle, representing less than 0.5% of the *global warming* for each formulation of tray. It is interesting to note that the end of life was more important for PP than for bioplastics, with end of life accounting for 26% and 2% of the total burden, respectively. This was mainly attributed to the incineration process. Incineration was more favorable for bioplastics and biocomposites because the carbon released was biogenic, unlike that from fossil-based plastics. The landfilling contribution to *global warming* was low, representing less than 5% of the PP end of life impacts, because PP was not assumed to be decomposed in the landfill. It must be noted that recycling of PP is an empty process because of the cutoff at recycling, meaning that the recycling benefit and costs are allocated to the production of new PP material.

In the present study, it was assumed that all the plastic wastes were managed without littering, but in reality, a non-negligible proportion of plastic waste ends in nature. In the world since 1950, 79% of plastic waste has accumulated in landfills or the natural environment (Geyer et al. 2017). Long-term impacts such as the accumulation of microplastics in the environment are currently not taken into account in LCA or only taken into account via unconnected tabulation of microplastic generation potential (Lee et al. 2014). Thus, some of the benefits of using bioplastics that fully biodegrade in natural conditions, relative to those that do not, are not quantified nor included in the analysis. This is particularly relevant for PHBV, which, unlike PLA, is fully biodegradable in soil and does not require industrial composting (Hermann et al. 2011). Furthermore, gas emissions from petrochemical polymer degradation, which have recently been demonstrated to produce methane and ethylene emissions under sunlight conditions in both water and air, are also not accounted for in LCA (Royer et al. 2018).

The nutrient contents of bioplastics (e.g., nitrogen and phosphorus) are so small that the benefit for reducing fertilizer use can be ignored. However, the sequestration of carbon in soil and the soil improvement properties are potential benefits of organic compost (Kim et al. 2008). Nevertheless, these are difficult to quantify and are considered outside of the scope of the present work.

3.2 Effect of the incorporation of ViSh fillers on the environmental performance of trays

A composite is the combination of two components: a matrix that constitutes the continuous phase, viz. PHBV, PLA, or PP in the present study, and fillers that corresponds to the dispersed phase, viz. ViSh particles in the present study. The *global warming* impact for 1 kg of material is displayed in Fig. 4 for the 4 possible constituents of composite materials. It was readily apparent that ViSh fillers exhibited a lower impact (0.26 kg CO_2eq/kg) than the polymer matrices (3.47, 3.58, and 2.29 kg CO_2eq/kg for PHBV, PLA, and PP, respectively). The ViSh global warming impact was nearly 9 times smaller than that of the PP matrix. This was due to the advantage of using agricultural residues that only required transport, drying, and milling.

Figure 5 shows how the *global warming* impact was affected by an increasing filler content in biocomposites. Similar figures for the other midpoint impact categories are available in SI. Through this assessment, a decreasing burden of the composite with increasing filler content was observed. Thus, the incorporation of ViSh appeared to be beneficial concerning *global warming.* It is worth noting that the production of composites required an additional compounding step and that the density of ViSh was 50% greater than that of PP, i.e., 1.36 g cm^{-3} for ViSh compared with 0.91 g cm^{-3} for PP. The burden incurred by the compounding step was noticeable for composites with very low filler contents. As the production of biocomposites induced an additional use

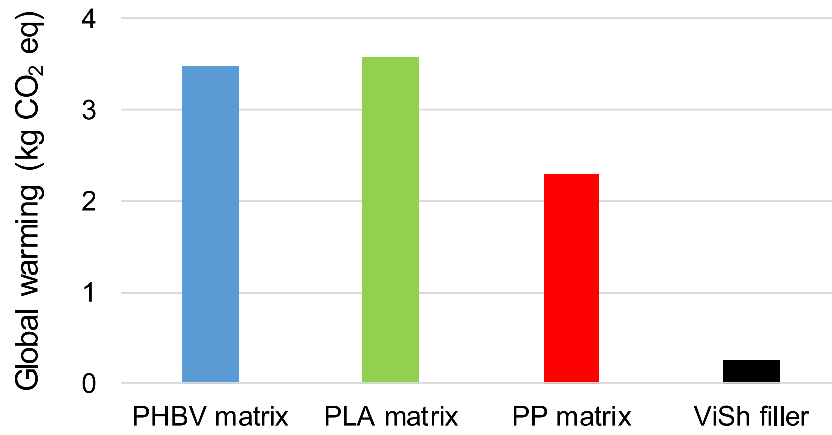


Fig. 4 Global warming impact (kg CO_2eq/kg) of 1 kg of each composite component

of energy, in all cases, composite with 1 vol% of ViSh had a higher *global warming* impact than respective virgin polymer matrices. The negative impact of the additional compounding step should be thus compensated for by the incorporation of increasing contents of ViSh particles in the polymer matrix. The magnitude of the decrease in impacts varied depending on the matrix type. For PHBV, PLA, and PP, the slope was respectively 1.00, 1.05, and 0.68 mg CO_2eq/%ViSh. Thus, the use of ViSh was beneficial from 5.5 vol% for PHBV and PLA, whereas the ViSh benefit in PP was first observed for a volume filler content of 20.0 vol%. PHBV-based composites had a lower contribution to *global warming* than 100% virgin PP tray, starting from a PHBV matrix with ViSh content of 44 vol%. However, this filler content is too high to be considered realistic, when taking into account the processability of the materials and their resulting mechanical properties. *Global warming* of PP-based composites was higher than PHBV-ViSh composites, only when reaching a ViSh content of 98.5 vol% and higher, which was of course a non-realistic formulation.

The filler content from which the addition of ViSh in the composite resulted in a benefit for all impact categories is displayed in Fig. 6. PHBV and PLA displayed similar results; the incorporation of ViSh improved the environmental impacts for all the categories except for *ionizing radiation*. If *ionizing radiation* was to be used as a single score indicator, then biocomposites would never exhibit lower impact than 100% virgin plastic trays because of the electricity needed for the milling, drying, and compounding steps of ViSh. The high *ionizing radiation* impact is mainly due to the French electricity mix, which includes a large share of electricity produced from nuclear power. In the case of PP, PP-based composite trays can be better than 100% PP trays in 10 of the 18 midpoint impact categories. The ViSh burden was higher than PP matrix in 4 midpoint impact categories, so accordingly the composite exhibited greater impacts in *strastospheric ozone depletion*, *ionizing radiation*, *land use*, and *mineral resource scarcity* than virgin PP. Similarly, the compounding step was responsible for the higher impact in *water consumption* and *terrestrial ecotoxicity*. Finally, the increased *freshwater and marine eutrophication* burden was due to the end of life of the composite. The black dashed line in Fig. 6 represents the limit of acceptable filler content of 30 vol% in the composite to ensure the functional unit. Thus, *freshwater and marine*

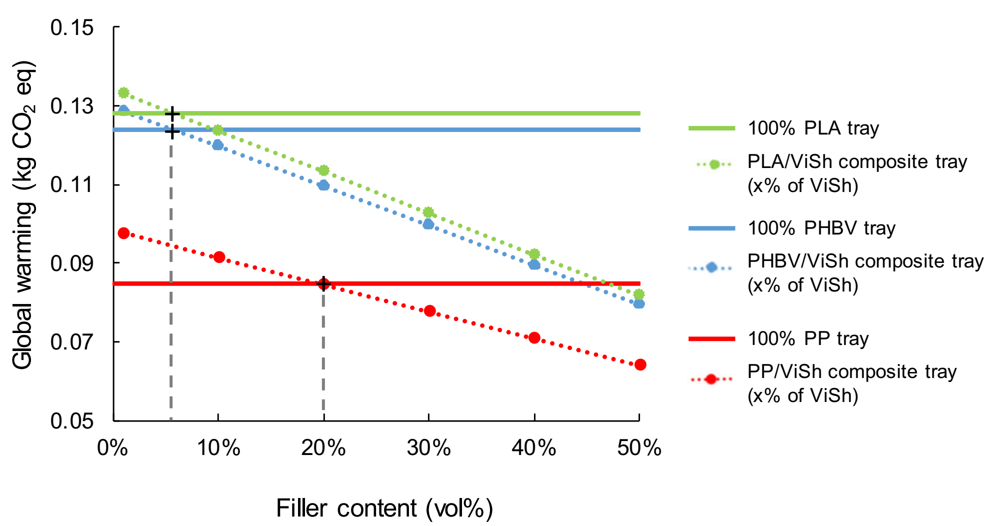


Fig. 5 Global warming impact (kgCO_2eq) as influenced by the filler content (vol%) for composite trays

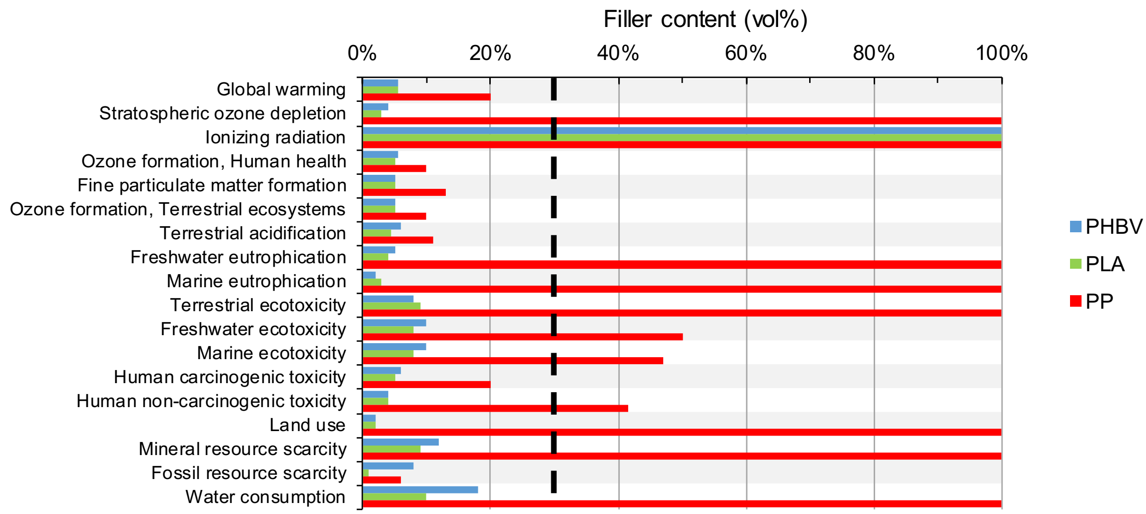


Fig. 6 Filler content (vol%) from which a composite tray results in lower environmental impacts than a 100% virgin plastic tray for each assessed impact category. The black dashed line represents the physical limitation of filler content (30 vol%) in the composite to ensure the functional unit. When bars reached a filler content of 100%, no benefit can be realized by the addition of filler

ecotoxicity and *human non-carcinogenic toxicity* were other impacts that PP-based composite could not improve relative to virgin PP.

According to results presented in Fig. 5 and Fig. 6, it could be concluded that increasing the ViSh filler content in the composites as much as possible, while respecting the restrictions set by material properties, was globally the best for the environment for all biocomposites. However, for PP, the inclusion of ViSH presents a case of burden shifting that would require more interpretation in order to determine overall environmental impact.

The environmental performance of composite trays filled with 30 vol% of ViSh particles was assessed in detail (Fig. 7). The 100% virgin PP tray was also added as reference. As previously described in Section 3.1, results were largely influenced by the nature of the matrix, mainly due to differences in density. PLA composites exhibited the highest environmental impact except for *ionizing radiation*, *terrestrial ecotoxicity*, *human non-carcinogenic toxicity*, and *land use* where PHBV exhibited the worst impacts. As expected, PP-based materials exhibited the highest impacts concerning *fossil resource scarcity*.

As shown on Fig. 8, *global warming* impacts of trays with 30 vol% ViSh fillers were significantly lower than those of trays made from 100% virgin plastics. This was in line with a previous study on the production of biocomposites with wheat straw (EcoBioCAP 265669 2013). The contributions were divided in three categories: (i) raw materials for matrix and ViSh fillers, (ii) processing for compounding and injection steps, and (iii) use and the end of life. The incorporation of 30 vol% of fillers reduced the *global warming* burden of the raw materials by 25% relative to a 100% plastic tray. Moreover, the end of life impacts were also reduced for bioplastics. In the case of a PP-based composite, PP could not be considered recyclable anymore, due to the presence of ViSh filler, inducing a slight increase of the EoL impact. Furthermore, the higher density of the composite materials relative to the pure plastics resulted in higher impacts from the injection molding step. And, the addition of ViSh came with an additional step of compounding, which had a relatively low impact compared with the injection molding process, as it represented 20% of the burden of the processing. Thus, the incorporation of 30 vol% of ViSh in trays reduced their *global warming* effects by 19.6%, 19.9%, and 8.5% for PHBV-, PLA-, and PP-based trays, respectively.

3.3 Identification of the hot spots

3.3.1 ViSh filler production: contribution of each step to the environmental impact

The main contributor to the environmental impacts of ViSh particles was the milling steps (Fig. 9). Milling represented 72% of the *global warming* impact, followed by the drying steps, with a contribution of 22%. The most burdensome type of milling was coarse milling, though there was no impact for *ionizing radiation* because the energy came from diesel fuel. This was contrary to electricity-powered cutting and fine millings. The fine milling step caused more impacts than cutting milling because more energy (electricity) is needed to get micrometric particles than millimetric particles. This should be expected, as total energy consumption increases as the particle size decreases regardless of milling equipment type (Mayer-Laigle et al. 2018). The final drying step also consumed energy, but in the form of heat from steam in the

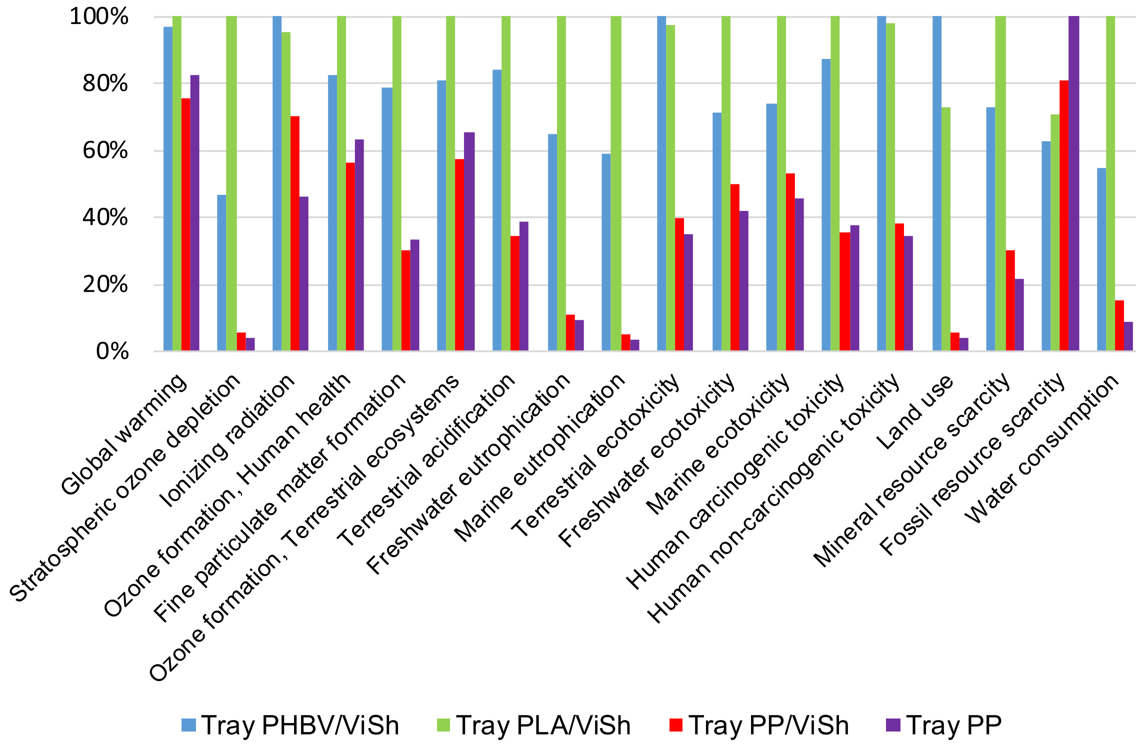


Fig. 7 Environmental impact of composite trays filled with 30 vol% of ViSh fillers (all impact categories)

chemical industry, which explained the low impact value in the *ionizing radiation* category.

The impact of ViSh transport was low in all the categories because it was assumed that the production of trays took place in the same region (Languedoc-Roussillon) as the generation of ViSh, allowing for short transportation distances.

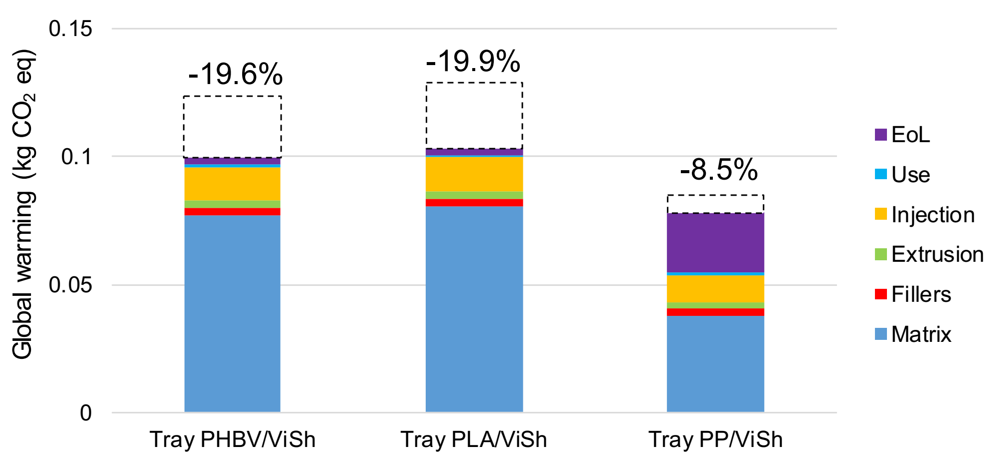
Air drying only exhibited environmental burdens in one impact category, since it only required space to spread the vine shoots on the ground without the help of machinery. Thus, impacts from this step only appeared in the category land use, representing 56% of the land use from ViSh production.

3.3.2 Polymer/ViSh (30 vol%) composite tray production: contribution of each step on the environmental impact

The analysis of the biocomposite burden clearly showed the strong contribution of the components of the composite and especially the matrix (Fig. 10). The contributions of PLA are not shown in Fig. 10 to increase clarity and because the results were very similar to those of PHBV composites.

For PHBV-based composites, the production of the polymer matrix was the largest contributor, 15 midpoint impact categories, ahead of the end of life (*freshwater and marine*

Fig. 8 Global warming impact of trays with 30 vol% filler. The percentages above the bars indicate the reduction of the impact compared with trays without ViSh filler



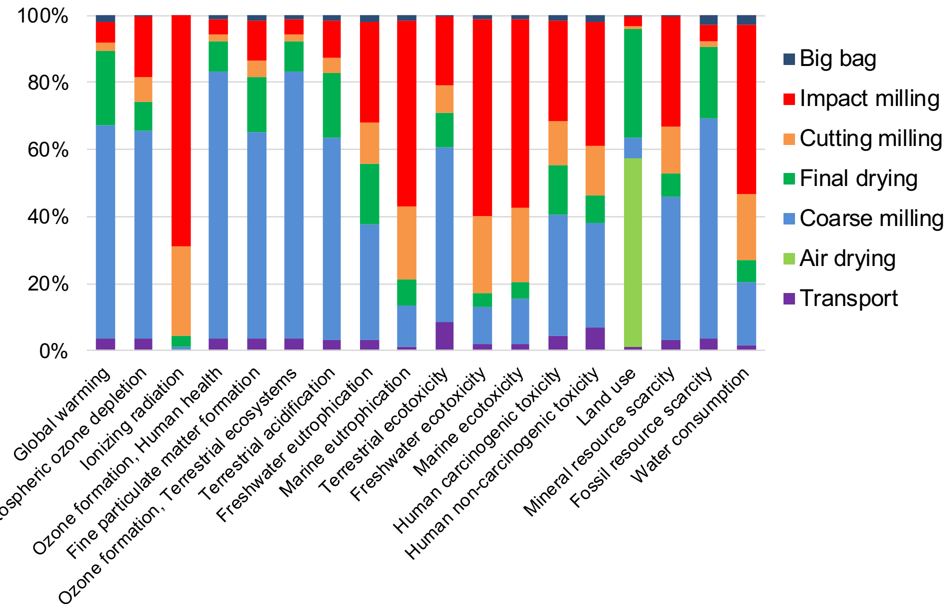


Fig. 9 Contribution for ViSh filler production

ecotoxicity) and the injection molding (*ionizing radiation*), in that order. The main contributions to the environmental burden in the production of PHBV are the large requirement for energy, in particular steam, and the use of sugar from sugar cane. In the case of PP-based composites, results were more balanced with 9 categories dominated by production of the matrix, 4 by injection molding or end of life, and 1 by compounding (*water consumption*). When comparing *global warming* potential, the production of the polymer matrix caused the largest contribution to environmental impact for the composite trays. The *global warming* impacts associated with polymer production outweighed those from the filler, manufacturing or end of life. The high contribution of the end of life step in the categories *freshwater and marine ecotoxicity* was mainly due to the landfilling.

As expected, *ionizing radiation* impacts were mainly due the manufacturing steps: injection molding and compounding. These processes required electricity. In the case of PHBV composite, *land use* impact was largely explained by the need of sugar cane that is used as carbon source for the production of the matrix.

3.4 Economic vs environmental balance analysis

The price of the different trays was estimated from data given by industry (Table 4). From an economic point of view, the

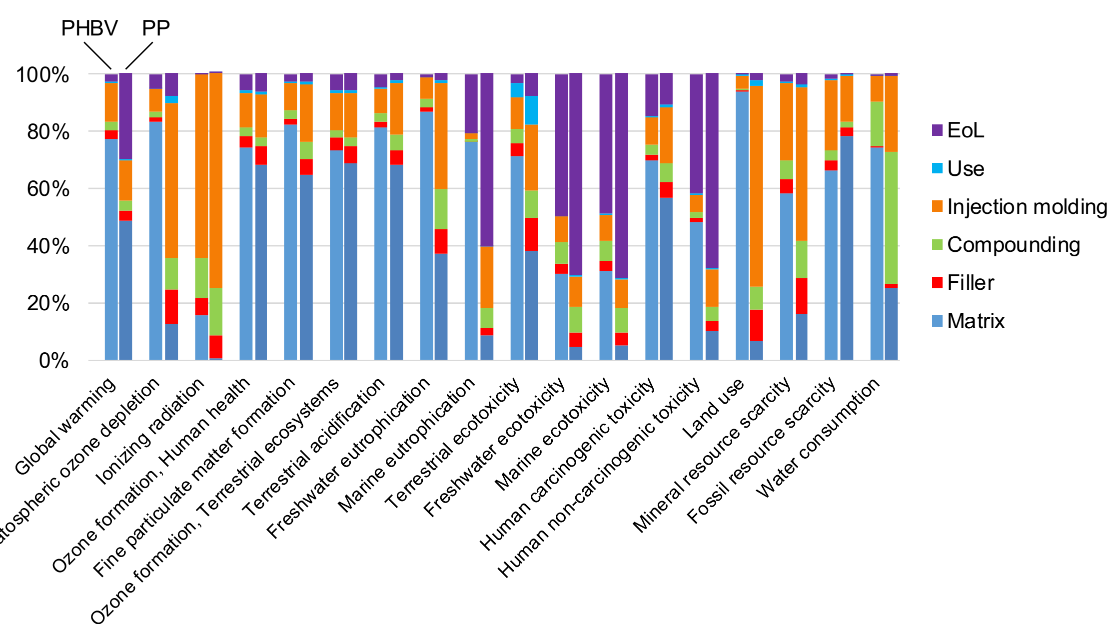


Fig. 10 Contribution for PHBV and PP-ViSh (30 vol%) composites

Table 4 Price of the studied composite trays. ViSh is 0.30 €/kg (*Vitis Valorem*, *ADEME*), the compounding is 0.04 €/kg (CT-IPC), and the injection molding is 0.03 €/p (Fürstplast)

	Price (€/t)	Price 100% plastic tray (€/100p)	Price 30 vol% ViSh filler tray (€/100p)	Reduction of the price due to 30 vol% of ViSh filler (%)
PHBV	7750[a]	26.95	20.11	− 25.4
PLA	2800[b]	11.73	9.46	− 19.4
PP	1240[c]	6.94	6.10	− 12.0

[a] NaturePlast, grade PHI 002, 2019

[b] NaturePlast, grade PLI 003, 2019

[c] French customs department, 2017

incorporation of 30 vol% of fillers reduced the price of a PHBV tray by 25.4%, and to a lesser extent in the case of PP (12.0%) because the price of raw PP is much lower than PHBV (Table 4). It is interesting to note that the injection molding accounted for a large share of the price, ranging from 12% for 100% PHBV trays to 50% for PP-based composite trays. On the contrary, in the case of composite materials, the additional price of compounding was almost negligible. This resulted in a factory price of final trays that was not only driven by the price of raw materials. Thus, the addition of ViSh in trays reduced the final price but not as much as expected according to the price of raw materials. There are two reasons for this: the price of injection molding, which was constant, and the density of ViSh was higher than for the plastics.

4 Conclusion

This study assessed the environmental impacts of composite trays made of PP, PLA, or PHBV, and increasing content of ViSh particle filler, based on a comparative life cycle assessment (LCA). It was shown that bioplastic matrices, i.e., PLA and PHBV, which are considered to be eco-friendly, displayed higher environmental impacts than fossil-based polypropylene. This result should be tempered by the fact that long-term impacts such as plastic accumulation are not considered and that the production of bioplastics is still at a much lower level of technological development. In the case of PHBV, the only truly biodegradable bioplastic among the three studied, it is expected that production processes will be optimized, in such a way to decrease their environmental impacts. It is therefore difficult to draw a general conclusion about the environmental efficiency of bioplastics compared with conventional plastics due to the expected evolution of the bioplastic technologies. As described by Yates and Barlow in a critical review on biopolymers (Yates and Barlow 2013), it is complex to compare their environmental impacts with other studies for different reasons: updated eco-profiles, feedstocks used, sources of energy, etc. There is currently no factor that quantifies the effect of plastic debris on biodiversity (Woods et al. 2016). The biodegradability of PHBV can thus not be assessed in the LCA framework. However, there is ongoing research on this issue (for example, the Marilca initiative supported by the Life Cycle Initiative of the UN Environment (Boulay et al. 2019)). One can only wonder how the conclusions of this work will change when such data become available. The interest of a biodegradable material, compared with a non-biodegradable material that is recyclable may seem low from a short-term life cycle analysis point of view. But, this perspective neglects the fate of the recycled material which, after a few cycles, will eventually be released into the environment, as the recycling of plastic, whether closed short loop or long loop, is limited in time.

The incorporation of increasing contents of ViSh particles in plastic trays resulted in a reduction of environmental impacts despite the additional processing steps required to produce ViSh fillers and the higher density of ViSh compared with the three polymer matrices under consideration. Trays with a higher filler content are therefore heavier requiring that more matter be processed. Despite that fact, this study illustrated the interest of using agro-residues in composites. Concerning global warming, composite trays had less impact than virgin plastic trays from 5 vol% for PHBV or PLA and from 20 vol% for PP. Regarding PHBV, the only biodegradable polymer in natural conditions in this study, the price and the impact on global warming are reduced by 25% and 20% respectively when 30 vol% of ViSh are added. Should the maximum filler content of 30 vol% be increased, there would be even greater potential to reduce the environmental impacts.

Thus, it can be concluded that, if the goal is environmental sustainability while avoiding microplastic accumulation, the majority research efforts should be devoted to the optimization and scale up of bioplastic production, PP production being already optimized. The use of cleaner energy would also help to achieve this goal while additionally reducing the impact of the injection molding step. Finally, the end of life should be also improved by increasing recycling for PP, ensuring separate collection for composting of PLA, and home composting for PHBV.

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