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Transforming Oleochemical production: enzymatic solutions for a low-carbon future

Abstract question: Does enzymatic catalysis lead to more sustainable chemicals production? A life cycle sustainability assessment of isopropyl palmitate.

Pieter Nachtegaele [1], Ozan Kocak [1] , Yblin Roman Escobar [1] , Jordy Motte[1], Dries Gabriels [2], Leopold Mottet [2] and Jo Dewulf [1]

[1] Ghent University ; [2] Oleon

In this work, a comprehensive Life Cycle Sustainability Assessment (LCSA) is performed assessing the environmental impact of switching from chemical to enzymatic catalysis for the esterification of Isopropyl palmitate (IPP). A dedicated LCSA methodology with a common goal, system boundary and life cycle inventory is presented. A 7 to 13% reduction in environmental impacts was found due to less hazardous waste formation, lower feedstock consumption and reduced steam usage. From a social and environmental perspective, upstream impacts linked to palmitic acid and isopropyl alcohol production should be addressed.

1. Introduction

Today's economy – from food, mobility, health to electronics – depends on the products of the chemical industry. Chemicals are also important building blocks in low-carbon, zero pollution and energy- and resource-efficient technologies, vital in finding new solutions for the green transition of our economy and society.[1] However, the sector also stands as the leading industrial energy consumer, and is associated with chemical pollution of soils, air and water.[2] In the past decades, the chemical industry has been actively seeking more environmentally sustainable production methods. Enzymes, for example, are promising biocatalysts that can increase energy efficiency, improve chemical safety, reduce chemical waste production, and even reduce the chemical production costs.[3] Enzymes are proteins that act as selective biological catalysts. Today, they are already used in various industrial sectors such as pharmaceuticals, cosmetics and food.[4,5] While enzymatic applications in the chemical industry have increased in recent years, this has not yet led to a major commercial breakthrough, and the number of products remains limited.[6–9]

Esters are an important class of chemicals with a wide range of applications such as emollients, surfactants, and emulsifiers in food, cosmetic, and pharmaceutical products.[9] Esterification reactions in industry typically use chemical catalysts, like strong acids, e.g., p-toluene sulfonic acid, or metals, e.g., tin or zinc salts, which lack selectivity and require harsh reaction conditions and complex downstream product purification.[7,9] Alternatively, lipases can be used for catalysing esterification reactions.[8] In previous studies, lipases were used to synthesize various esters and were found to be promising due to mild reaction conditions and reduced hazardous waste formation.[9–11] The main drawbacks, however, are the high enzyme cost and

the difficulty of reusing lipases due to irreversible enzyme inactivation.[6] Immobilization of lipases, by attaching them to a solid phase or support, has been an important step to enabling multiple reuses of lipases.[12,13]

Isopropyl palmitate (IPP) is an oleochemical ester, commercially used as an emollient in cosmetics, healthcare products and lubricants due to its good absorption characteristics.[14] IPP is typically synthesized by the esterification of palmitic acid (PA) and isopropyl alcohol (IPA) using a chemical acid catalyst such as sulfonic acid.[15] Various studies have successfully used lipases for the production of IPP in a lab-scale batch configuration.[8,14] Furthermore, kinetic models have been studied and the optimum reaction conditions have been proposed.[16,17] However, to the authors' knowledge, enzymatic IPP production at pilot or industrial scale has not been reported. Although switching from chemical to enzymatic catalysis is largely perceived as sustainable, it is essential to measure the environmental impact before implementation on an industrial scale.[8] Life Cycle Sustainability Assessment (LCSA) is a comprehensive framework for evaluating the three pillars of sustainability for a product, process, or service throughout its life cycle.[18] However, to the authors' best knowledge, no environmental sustainability assessment for enzymatic IPP production, and no comprehensive LCSA comparing enzymatic and chemical production in general, has been reported in the literature.

In this study, a comprehensive LCSA is presented assessing and comparing the environmental (LCA) performance of enzymatic and chemical catalysis for the esterification of IPP. This study presents for the first time a sustainability assessment of IPP production based on pilot-scale data. In addition, it presents the first detailed LCA of this process. A contribution analysis was performed to identify the main environmental sustainability hotspots in the enzymatic process. A sensitivity analysis was conducted to investigate the importance of reusing enzymes and sustainable feedstock sourcing.

2. Materials and Methods

The applied LCSA methodology (Fig. 1) for comparing chemical and enzymatic IPP production followed the four phases of the ISO14040s framework for Environmental LCA: (i) Goal and Scope definition, (ii) Life Cycle Inventory (LCI) (iii) Life Cycle Impact Assessment (LCIA) and (iv) Interpretation.[19] In this study, a common goal and scope definition was defined and a common LCI was collected to streamline the environmental, social and techno-economic assessments. The LCA in this study has been conducted in accordance with ISO 14040 and ISO 14044 standards.[19,20]

Goal and scope definition

The goal of this study was to assess the environmental life cycle sustainability of the production of Isopropyl Palmitate (IPP) via enzymatic esterification and identify relevant hotspots that can affect its sustainability performance. In addition, the study aimed to compare the sustainability of enzymatically produced IPP to conventional chemically produced IPP. For all assessments, the functional unit was defined as one kilogram of IPP produced via chemical or

enzymatic production with the same functionality and a cradle-to-gate system boundary was used, meaning that the processes from resource extraction to the factory gate were considered as presented in Fig. 2.

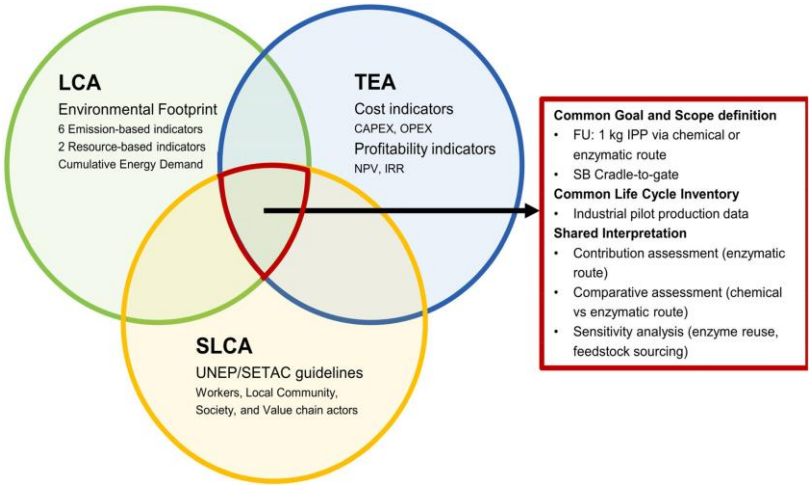

Fig. 1 Life cycle sustainability assessment methodology for comparing enzymatic and chemical production of IPP. FU = functional unit. SB = system boundaries. NPV = net present value. IRR = internal-rate-of-return. OPEX = operational expenditures. CAPEX = capital expenditures.

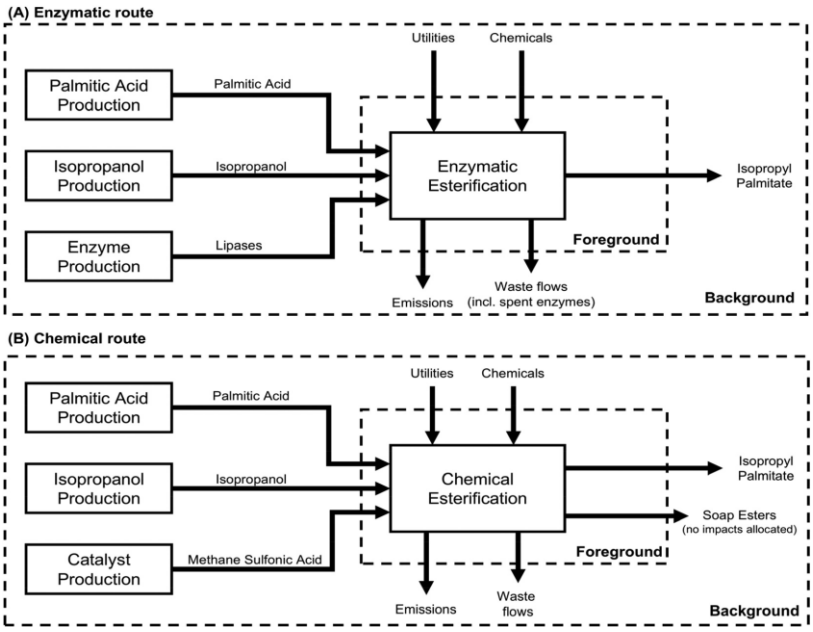

Fig. 2 Simplified flow diagrams of the production of IPP via (A) enzymatic route and (B) chemical route.

In the enzymatic route, IPP is produced via a lipase-catalysed, solvent-free process. The lipase-catalysed esterification process includes vessel preparation, esterification, enzyme recycling, and post-treatment (deodorization and filtration). The esterification process requires

PA, IPA and enzymes (lipases) and takes place at 60 °C. PA is derived from crude palm oil through a series of steps including hydrolysis, distillation, and fractionation. Isopropyl alcohol is a fossil-based chemical, and it is typically produced by hydration of propene or by gas fermentation technology using steel mill off-gas. Enzymes used in the analysis, Novozym® 435, are an immobilized form of Candida Antarctica lipase B, and produced by Novozymes A/S.[21] The immobilized enzyme catalyst is isolated/fixed in a separate column in the installation. Since the enzyme column is only used for esterification, multiple use of enzymes is possible without the need for harsh cleaning between batches. Based on pilot testing, it is assumed that these enzymes are reused 20 times. Aside from these materials, the enzymatic route also includes the consumption of utilities (electricity, steam, and nitrogen), for esterification and post-treatment, and filter aids for filtration. The removed isopropyl alcohol, waste filter aids and spent enzymes are incinerated as chemical waste. The chemical route takes place in a

stirred reactor tank at the same location. The route includes esterification, isopropyl alcohol removal and post-treatment (distillation, neutralization and washing, deodorization). In the chemical route, methane sulfonic acid (MSA) was used as an acid catalyst in the esterification, along with PA, IPA and auxiliary chemicals (e.g. NaOH). Esterification takes place at 130 °C. A neutralization step produces soap esters as a by-product. Because of the limited amount and low value, no impact is allocated to this by-product. The relative batch time of enzymatic esterification compared to chemical esterification is 4 : 1.

Life cycle inventory

Primary data for the enzymatic and chemical route was collected from respectively Oleon's pilot facility and the industrial batch process at their production site in Oelegem, Belgium. The aggregated LCI is available in ESI S2. Due to confidentiality of Oleon's industrial process data, the detailed LCI, giving the mass and energy balance of the chemical and enzymatic process, is not included. However, basic process information is shared in Table 1.

Table 1: Operational conditions of enzymatic and chemical routes

	Enzymatic route	Chemical route
Catalyst	Novozym® 435	Methane sulfonic acid
Temperature (°C)	60	130
Pressure (atm)	1	1
Post-treatment	Deodorization, filtration	Distillation, neutralisation and washing, deodorisation
Relative batch time	4	1

Fig. 2 illustrates the differentiation between the foreground and background systems. For the background data, ecoinvent database version 3.9.1[22] was utilized. 'Market for' activities were selected if available to ensure the inclusion of transportation to the production site. For the PA production, palm oil produced in Malaysia was considered as feedstock. Additionally, the transportation of palm oil from Malaysia to the plant in Belgium was included. The conversion of palm oil to PA was modelled using internal Oleon data for fatty acid hydrogenation and fractionation processes, as described in Nachtergaele et al. (2019).[23] A life cycle inventory on

lipase production was provided by the supplier company Novozymes A/S. The LCI of enzyme production is included in the aggregated LCI in ESI (S2), however, the detailed LCI cannot be made available due to industry confidentiality.

Environmental life cycle impact assessment

The LCA in this study has been conducted in accordance with ISO 14040 and ISO 14044 standards.[19,20] For LCA, the maintenance and infrastructure aspects were excluded. In the impact assessment phase of this study, the Environmental Footprint (EF) v3.1 method[24] was used. From the 16 impact categories included in this method, six emission-based impact categories were selected according to the recommendations of the Life Cycle Metrics for Chemicals Products.[25] In addition, two resource-based impact categories were evaluated, namely land use and water use. Furthermore, primary energy consumption was analysed by assessing the Cumulative Energy Demand (CED).[26] SimaPro® software version 9.5[27] was used for conducting the LCIA.

Interpretation

Within the scope of this study, a comparative assessment of the environmental impacts of the enzymatic and chemical routes was performed. Furthermore, a contribution analysis was carried out to identify sustainability hotspots in the enzymatic process. For LCA, an uncertainty analysis was included to assess the robustness of the results to uncertainty in the background data. The uncertainty assessment was performed by a Monte Carlo analysis of 10 000 runs. Finally, sensitivity analyses were performed to investigate the effect of enzyme reuse on environmental and economic sustainability and feedstock sourcing on environmental and social sustainability of enzymatic IPP.

3. Results and discussion

Environmental impact of IPP production

The results of the contribution analysis for the enzymatic production route are listed in Table 2. PA production is the main contributor to all impact categories. For climate change, the contribution of this feedstock amounts to 81%, primarily due to land-use change from deforestation, high energy consumption and waste generation during palm oil cultivation. Deforestation and related biodiversity loss are also reflected in the high contribution of palm oil within the land use impact category (99%).[28] The other feedstock, IPA, also has a significant contribution for most assessed impact categories, particularly water use (25%), cumulative energy demand (17%), acidification (13%) and climate change (9%). The primary raw material for IPA production is propylene, which is typically derived from fossil resources through energy-intensive processes.[29] Producing IPA from alternative feedstocks is a promising route to reduce the environmental impact of this input.[30] For example, a recent study on the production of IPA from industrial waste gas feedstocks through fermentation reported a negative cradle-to-gate carbon footprint of −1.17 kg CO2 eq. per kg of produced IPA. The negative value is acquired by considering the avoided off-gas emissions.[31] In contrast to both feedstocks, enzyme production has only a minor contribution on the total environmental impact

of IPP production for all examined impact categories. It is important to note that in the base case, the assumption is made that enzymes are reused 20 times. Notably, for water use, enzymes contribute 8% to the total impact, primarily attributed to the fermentation and purification stages in enzyme production.[32,33] Utilities such as electricity, steam, and nitrogen have relatively low contributions across all analysed categories. While waste treatment shows a low impact in categories such as acidification (3%) and cumulative energy demand (2%), its relative impacts are higher in the ecotoxicity (10%) and human toxicity (12%) categories. It is important to note that in this study, the wastewater is classified as hazardous waste due to the presence of isopropanol. In a full-scale industrial plant, it is expected that improvements in liquid waste treatment will reduce this impact.

Table 2 Environmental impacts of 1 kg IPP production via enzymatic route for the selected impact categories

		Feedstock supply			Esterification			
Impact category	Unit	PA	IPA	Catalyst supply Enzyme	Chemicals	Utilities	Waste	Total
Acidification	mol H+ eq	1.4×10^{-2}	1.7×10^{-3}	1.90×10^{-4}	7.2×10^{-5}	1.6×10^{-4}	4.2×10^{-4}	1.6×10^{-2}
Climate change	kg CO_2 eq.	3.9×10	4.3×10^{-1}	3.00×10^{-2}	2.0×10^{-2}	1.0×10^{-1}	3.6×10^{-1}	4.8×10
Ecotoxicity, freshwater	CTUe	4.5×10^{1}	2.7×10^{-1}	1.30×10^{-1}	2.3×10^{-1}	8.0×10^{-2}	5.3×10	5.1×10^{1}
Eutrophication, freshwater	kg P eq	1.6×10^{-4}	3.5×10^{-6}	1.00×10^{-6}	2.6×10^{-7}	1.6×10^{-6}	7.4×10^{-6}	1.8×10^{-4}
Human toxicity, cancer	CTUh	1.4×10^{-9}	3.4×10^{-11}	1.70×10^{-11}	4.6×10^{-11}	6.8×10^{-12}	2.0×10^{-10}	1.7×10^{-9}
Land use	Pt	8.8×10^{1}	1.2×10^{-1}	3.30×10^{-1}	3.0×10^{-2}	3.3×10^{-1}	8.0×10^{-2}	8.9×10^{1}
Photochemical ozone formation	kg NMVOC eq	8.5×10^{-3}	1.8×10^{-3}	8.50×10^{-5}	4.9×10^{-5}	1.7×10^{-4}	3.8×10^{-4}	1.1×10^{-2}
Water use	m^3 depriv.	2.8×10^{-1}	1.4×10^{-1}	4.00×10^{-2}	0.0×10	4.0×10^{-2}	4.0×10^{-2}	5.5×10^{-1}
Cumulative Energy Demand	MJ	8.2×10^{1}	1.4×10^{1}	4.70×10^{-1}	1.5×10^{-1}	3.5×10	1.8×10	1.0×10^{2}

Fig. 3 presents the relative environmental impacts for IPP production for the enzymatic route (ER) and chemical route 13% compared to the chemical route, depending on the considered impact category. The reduction is most prominent for climate change and photochemical ozone formation, with reductions of 13%. It is possible to attribute this reduction to the triple advantage observed for the enzymatic route, namely (1) reduced chemical waste (2) higher yield, which results in a lower impact from the feedstocks PA and IPA, and (3) reduced steam consumption (part of utilities) due to the lower process temperature and easier downstream processing. The uncertainty analysis (see ESI S1†) showed that the enzymatic route consistently (100%) scores better than the chemical route for the indicators acidification, climate change, eutrophication (freshwater), land use, photochemical ozone formation and water use, while the uncertainty in the background data may result in reversed results for ecotoxicity (freshwater), human toxicity (cancer) and land use.The uncertainty analysis (see ESI S1†) showed that the enzymatic route consistently (100%) scores better than the chemical route for the indicators acidification, climate change, eutrophication (freshwater), land use, photochemical ozone formation and water use, while the uncertainty in the background data may result in reversed results for ecotoxicity (freshwater), human toxicity (cancer) and land use.

The effect of enzyme reuse

The importance of reusing enzymes for multiple batches was examined through a sensitivity analysis. Based on pilot testing, the base case was to use the enzyme column for 20 batches before reloading. The climate change impacts of IPP when using enzymes for 1, 10, and 25

batches are also presented in Fig. 4(A). In the base case the contribution of enzymes to the climate impact category was below 1%. In case of no reuse, the contribution increased to 12%, resulting in a climate change impact for the enzymatic route close to the chemical route. These results show the importance of reusing enzymes for multiple batches to achieve environmental sustainability gains. Using enzymes for 25 batches does not lead to a significant change compared to 20 batches in terms of environmental sustainability.

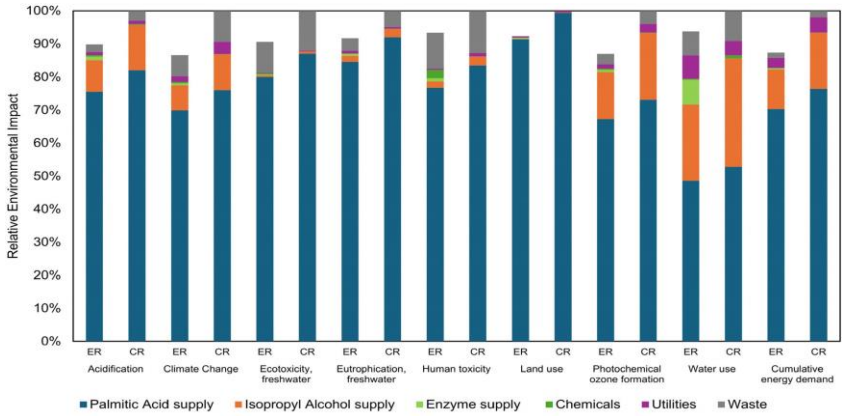

Fig. 3 Comparative life cycle impact assessment of the enzymatic route (ER) and chemical route(CR).

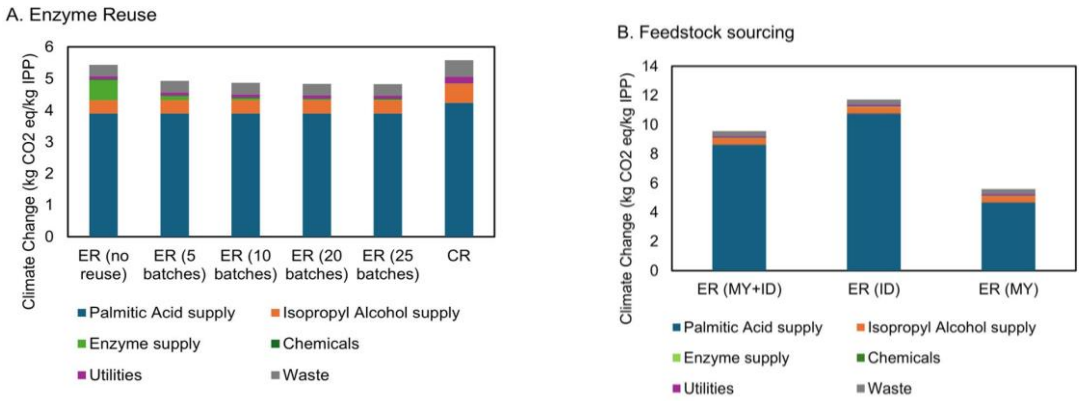

Fig. 4 Sensitivity analysis of the different scenarios to investigate the impact on climate change for (A) enzyme recycling and (B) different sourcing of palmitic acid. ER = enzymatic route, CR = chemical route. ER = enzymatic route, CR = chemical route. MY = Malaysia. ID = Indonesia

The effect of feedstock sourcing

The results of the hotspot assessment in Fig. 3 show the high contribution of palmitic acid, and more specifically palm oil cultivation, for all environmental impact indicators. The base case considered crude palm oil produced in Malaysia. The effects of changing the supply from Malaysia to Indonesia or a mix from both Malaysia and Indonesia were analysed. In the case of the mixed supply from both countries, the assumption was that 68.84% of palm oil was

supplied from Indonesia and 31.16% from Malaysia. For this assessment, the Agri-footprint database was used for palm oil from both Malaysia and Indonesia, as palm oil from Indonesia was not available in ecoinvent at the time of the assessment. Fig. 4(B) shows that sourcing palm oil from Indonesia rather than Malaysia would more than double the overall climate change impact. This demonstrates that the sourcing of palm oil has a significant impact on the environmental sustainability of IPP production. In both countries, conventional methods of palm oil production cause the destruction of carbon-rich forests and peatlands, contributing to global warming.[34] However, less deforestation occurred in Malaysia, both in absolute and relative terms, in previous decades.[35] The International Sustainability & Carbon Certification (ISCC) is an international certification system that covers various bio-based feedstocks and renewables, including palm oil. This certification ensures that the feedstock was not cultivated on land with high biodiversity or high carbon, thereby protecting against deforestation and indirect land-use.[36] To account for ISCC certification, the crude palm oil data used were modified by excluding the burdens of “land-use transformation” and “CO2 emission due to land transformation”.[23] Using ISCC certified palm oil as feedstock reduced the climate change impact for the enzymatic and chemical route to 1.[54] and 2.00 kg CO2 eq. per kg IPP, a reduction of respectively 68% and 64%. Due to the smaller overall impact of IPP production, the relative reduction of the enzymatic route on the climate change impact compared to the chemical route increased from 13 to 23%.

4. Conclusion

The sustainability of producing 1 kg IPP via enzymatic catalysis was compared to conventional chemical catalysis. It was found that the feedstocks, and specifically PA, were the main contributors to the environmental cost of IPP. Developments toward more sustainable palm oil cultivation and the production of bio-based IPA could therefore result in a significant reduction of the environmental impact of IPP. The comparative assessment showed that switching to enzymatic catalysis for IPP production reduced the environmental impacts between 7 and 13%, depending on the considered impact category. This was due to a triple benefit, being the production of less hazardous waste, lower feedstock consumption due to higher yield, and a lower steam consumption. The performed sensitivity analysis underscores the crucial role of enzyme reuse for environmental and economic sustainability. The proposed LCSA methodology provided clear guidance and insights on assessing and improving the life cycle sustainability of enzymatic catalysis for chemicals production. Using an LCSA methodology with a common goal and scope definition and life cycle inventory reduced the overall time for data collection and streamlined the interpretation. However, several limitations to the current methodology should be noted. Firstly, the methodology currently does not account for differences in scale when comparing enzymatic and chemical catalysis. Comparing technologies at low technology readiness levels (TRLs) with mature processes, which benefit from high levels of process integration and decades of optimization, may lead to an underestimation of sustainability gains.[37] Existing prospective sustainability assessment frameworks, e.g., Thonemann et al. (2020),[37] should be tailored for predicting the industrial scale LCI of enzymatic catalysis when still at lab or pilot scale (TRL 3–5). Secondly, the PSILCA database is currently suggested for

the SLCA. However, this database provides only sector and country specific data, lacking detailed information about the system under study.[29]

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