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Exergy-Based Efficiency and Renewability Assessment of Biofuel Production

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This study presents an efficiency and renewability analysis of the production of three biofuels: rapeseed methyl ester (RME), soybean methyl ester (SME) and corn-based ethanol (EtOH). The overall production chains have been taken into account: not only the agricultural crop production and the industrial conversion into biofuel, but also production of the supply of agricultural resources (pesticides, fertilizers, fuel, seeding material) and industrial resources (energy and chemicals) to transform the crops into biofuel. Simultaneously, byproducts of the agricultural and industrial processes have been taken into account when resources have to be allocated to the biofuels. The technical analysis via the second law of thermodynamics revealed that corn-based EtOH results in the highest production rate with an exergetic fuel content of 68.8 GJ ha^{-1} yr^{-1}, whereas the RME and SME results were limited to 47.5 and 16.4 GJ ha^{-1} yr^{-1}. The allocated nonrenewable resource input to deliver these biofuels is significant: 16.5, 15.4, and 5.6 MJ ha^{-1} yr^{-1}. This means that these biofuels, generally considered as renewable resources, embed a nonrenewable fraction of one-quarter for EtOH and even one-third for RME and SME. This type of analysis provides scientifically sound quantitative information that is necessary with respect to the sustainability analysis of so-called renewable energy.

Introduction

Currently, a number of "biofuels", that is, alcohols, esters, ethers, and other chemicals made from biomass, are in focus with respect to their substitution for fossil fuels. Two major types of fuels from agricultural crops are getting growing attention as biofuel for transportation purposes. First, via fermentation, ethanol can be obtained from a large number of crops: corn, sugar beet, sugar cane, and cellulosic materials. Second, from a second group of crops with a substantial fraction of triglycerides, such as rapeseed and soy, methyl esters are produced through chemical transesterification. These fuels, soybean methyl ester (SME) and rapeseed methyl ester (RME), have properties that are comparable to diesel fuel. Biofuels have a number of characteristics allowing labeling them as "sustainable fuels". Indeed, they are based on the ultimate renewable energy resource: solar irradiation. Simultaneously, they close the carbon cycle, making them CO_2 neutral.

With respect to an overall technical sustainability assessment of products, three major issues have to be considered (*1*−*5*). First, immediate environmental compatibility is important: extraction of renewable and nonrenewable resources and waste emissions during the product's life cycle have to be considered. Second, integration within the industrial ecological system through making use of waste materials instead of virgin resources, on one hand, and delivering products that are recoverable at the end of life, on the other hand, generates environmental benefits. Third, a higher efficiency of the overall production provides not only economic benefit but contributes to less resource consumption and reduced emissions. With these technical sustainability issues in mind, it is not straightforward to conclude that biofuels inherently contribute to sustainability. Although net carbon dioxide emission is expected to be reduced drastically when compared to fossil fuels, all gaseous, liquid, and solid emissions at the production stage should be considered, during both the agricultural crop production and the industrial biofuel production. Next, a number of nonrenewable resources are required for both the agricultural and industrial production stages so that the renewability degree cannot be 100%. Moreover, biofuels require agricultural land, which is then no longer available for fulfilment of other needs, such as food or feed production. Efficiency of the agricultural−industrial production chain is important: the less efficient, the higher the land requirements for the same fuel production. Finally, the assessment of biofuels is complicated by so-called joint production. Indeed, a number of crops not only result in biofuels after an industrial post-harvest conversion, but also are embedded within the industrial ecological system, being used as fodder or fertilizer. The same is valid for the industrial conversion of the crop product into biofuel: useful byproducts, such as glycerol and soy meal, are of value in the overall industrial ecological system. An accurate sustainability assessment of biofuels must take into consideration a suitable procedure to allocate agricultural and industrial input materials to the biofuels and the agricultural and industrial byproducts.

It is clear that metrics are required to assess the contribution of biofuels to sustainability through a thorough process analysis. Energy analysis has been criticized, since it only includes the quantity of energy, not its quality. According to the first law of thermodynamics, energy is indestructible, which means that energy is conserved in every process. Energy can only be converted into different forms. However, the quality of energy is consumed in each conversion. In exergy analysis, the quality of the energy is taken into account. Unlike energy, exergy is consumed in all real world processes as entropy is produced. In recent literature, the exergy concept has been shown to be a powerful tool in the sustainability assessment of technology (*1*−*9*) and in the analysis of bioenergy systems (*1*, *10*−*11*). The exergy concept is the thermodynamic tool that expresses the quality of energy: it is the amount of work that can be obtained from a system when it is brought in equilibrium with the environmental state, the so-called dead state. There is extensive information available in the literature about the exergy concept (*12*−*14*). Exergy and energy analysis results can be similar or different, depending on the studied processes. Processes with limited entropy production, for example, chemical conversions with limited heat production, may result in comparable energy and exergy results. Processes accompanied by substantial entropy production, for example, heat production out of chemicals or electricity, result in significant differences between the two types of analyses.

In technology assessment by exergy, it is obvious that the cumulative exergy consumption (CExC) is a key factor: it

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describes quantitatively the amount of resources, both renewable and nonrenewable, that has to be extracted out of the ecosystem in order to deliver a product. It immediately allows the calculation of the efficiency of a production chain. Finally, by the fact that it is able to quantify all types of materials on the same universal thermodynamic basis, whether they are input or output, it allows a straightforward procedure to allocate inputs to outputs.

The purpose of this work is to contribute to the sustainability assessment with respect to the renewability degree and efficiency of biofuel production. Three biofuels are compared: rapeseed methyl ester, soybean methyl ester, and corn-based ethanol.

Methods

Three production chains were quantified in exergy terms. The reference state of Szargut et al. (*14*) was considered with its reference temperature (298 K), pressure (1 atm), and composition. For water, freshwater was chosen to be the reference state. If not available, calculation of exergy values was performed by group contribution methods (*14*−*17*) from Gibbs energy of formation data (*18*) or by calculating lower heating value-to-exergy ratios (*14*, *19*). Solar irradiations were calculated by taking into account the geographical position of the different production sites and by making use of the solar energy engineering website of the University of Massachusetts, Lowell (*20*). Exergy of solar radiation can be calculated from the exergy-to-energy ratio being 0.933 (*14*). Cumulative exergy values were calculated by tracing back input materials to the raw materials extracted out of the ecosystem to deliver them. With respect to system boundaries, back-tracing was limited to levels 1 and 2 according to the system of Szargut et al. (*14*). This means that the whole production chain, including extraction, transport and storage of raw materials and the fabrication of intermediates (e.g., pesticides, steam) is taken into account. Capital equipment production has not been included; exergy for capital equipment has been shown to contribute in a limited way, usually below 5−10% (*14*).

Studied Materials

Rapeseed Methyl Ester (RME). The analysis of RME production is based on process data from refs *10*, *21*−*22*. Considered inputs for the rapeseed production were seeds, solar energy (Swedish conditions), burnt lime, nitrogen−phosphorus−potassium and calcium ammonium nitrate fertilizers, pesticides (glyphosate, deltamethrin, metazachlor, surface-active Lissapol, and manganese micronutrient Mantrac), and fuel consumption for the operation of agricultural machinery and transport. Outputs are rapeseeds and straw. At the industrial production with four substages, that is, pretreatment (drying, cleaning, and storage), extraction, refining, and esterification, considered input materials are rapeseeds, power consumption (electricity, steam, fuels), and chemicals (phosphoric acid, hexane, bleaching material, sodium hydroxide, hydrogen chloride, methanol, sodium methylate). Products are meal, RME, and glycerol. Quantities of products are 3000 kg rapeseeds and 1190 kg RME ha^{-1} yr^{-1}.

Soybean Methyl Ester (SME). The production process of SME is similar to RME production (*23*), with similar agricultural inputs and outputs. Product data are 2554 kg soybeans and 418.5 kg SME per ha and year (average based on 14 key soybean-producing states within the U.S.A.).

Corn-Based Ethanol (EtOH). The production route of corn (*11*) takes into account the agricultural production with use of seeds, pesticides, fertilizers, fuels, and solar energy (Italian conditions), being similar to rapeseed and soy production, delivering kernel and stover. The industrial stage, however, is different, since the main process is a biological fermentation preceded by milling and hydrolysis and followed by liquid/solid separation and distillation. These processes require energy and chemicals (enzymes, yeast, sodium chloride, calcium oxide, ammonia, denaturants). Products are ethanol and distiller's dried grains with solubles (DGGS). A production of 7600 kg kernel and 2330 kg EtOH ha^{-1} yr^{-1} were taken into account.

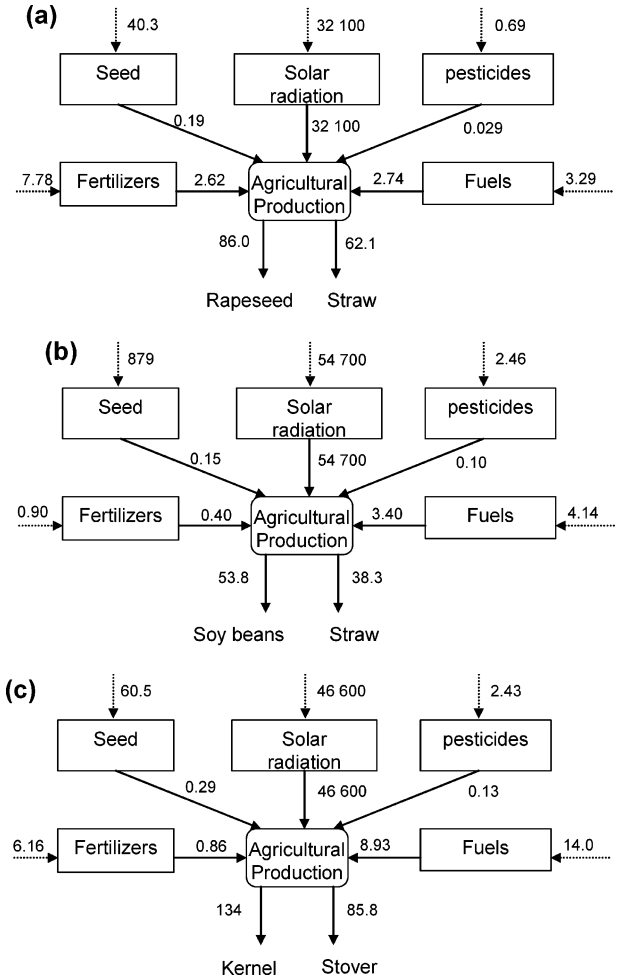

FIGURE 1. Overview of inputs and outputs related to 1 ha of agricultural production of biofuel resources. All data are in GJ of exergy ha^{-1} yr^{-1}. Data at dotted lines are the cumulative exergy extracted out of the natural environment to deliver the input materials. (a) Rapeseed; (b) soybean; (c) corn.

Results and Discussion

Agricultural Production Stage. In Figure 1, the agricultural production stages for rapeseed, soybean, and corn are represented with all exergetic inputs and outputs. It turns out that solar radiation is the major input source, accounting for more than 99.9%. In this sense, it can be stated that the agricultural products are largely renewable. On the basis of the huge solar energy input, the production is not efficient. Overall output-to-input ratios are limited to 0.46, 0.17, and 0.47% for the rapeseed, soy, and corn production, respectively. This is due to the nature of the photosynthesis process: both efficiency and period of operation (growing season) are limited, for example, when compared to photovoltaic electricity generation (*1*, *24*).

When the three production manners are compared, it turns out that corn production delivers the largest amount of exergy, 220 GJ ha^{-1} yr^{-1}, better than rapeseed (148) and soy (92). This is related to the fact that corn has a more

TABLE 1. Inputs and Outputs (GJ of Exergy) of the Conversion of the Production of 1 ha of Crops into Biofuels

	rapeseed	soybean	corn
Inputs			
agricultural product	86.0	53.8	134
fuels	0.17	2.97	0.32
electricity	0.46	0.69	2.85
steam	1.00	0.77	
chemicals	3.63	1.30	1.79
sum	91.3	59.5	139
Outputs			
biofuel			
RME	47.5		
SME		16.4	
EtOH			68.8
byproducts			
glycerol	2.10	1.31	
meal	32.7	37.3	
DDGS			43.4
sum	82.3	55.0	112

efficient biosynthesis route; it is a so-called C4 plant type. It has also to be noticed that byproducts straw and stover have an important share in the output, being very similar in the three cases: 42.0, 41.6 and 39.0% for rapeseed, soy, and corn, respectively. These byproducts either can be used as fodder or can stay on the field as fertilizer for the next crop. Hence, they have to be taken into account when inputs to the agricultural production have to be allocated.

Finally, the production manners can be assessed in terms of exergy breeding, similar to energy breeding. The energy or exergy breeding factor (BF_{en} or BF_{ex}) (*25*) and the net energy or exergy value (NEnV or NExV) (*26*) assesses how much renewable resources are bred from nonrenewable resources. To do so, renewable inputs have to be subtracted from the total input. Not only solar radiation has to be considered here, but one should also pay attention to the nature of the seeding material. The production of rapeseed seeding material, for example, is basically the rapeseed production scheme in Figure 1a, showing that for seeding material from 1 ha cultivation, solar input is overwhelming (32 100 GJ) when compared to the other inputs fertilizers, pesticides, and fuels (5.39 GJ), omitting the 0.19 GJ of seeding material. This means that the input for seeding material is more than 99.9% renewable in nature, thus allowing it to be considered as renewable. The same is true for soybeans and corn. In turn, the contribution of the seeding material to the total renewable input is negligible, since it is 3 orders of magnitude lower than the direct solar input for the three crops. Taking into account the nonrenewable fertilizers, pesticides, and fuels, it turns out that corn, rapeseed, and soy production show total nonrenewable inputs of 9.92, 5.39, and 3.90, with an output of 220, 148 and 92.1 MJ ha^{-1} yr^{-1}, respectively. This means that BF_{ex} values of 22.2, 27.5 and 23.6 are obtained for the respective production routes. However, it should be taken into account that inputs for agriculture require an industrial production stage before they can be supplied, for example, chemical manufacturing of pesticides and fertilizers. If one aims at a global chain assessment, one should make the ratio between the exergy embodied in the resources extracted out of the ecosystem that goes into the agricultural production through these industrial supplying chains: the cumulative exergy consumption. With a global cumulative nonrenewable input of 22.6, 11.8, and 7.5 GJ, respectively, it turns out that overall, BF_{ex} values decrease to 9.7, 12.6, and 12.3 for corn, rapeseed, and soy, respectively.

Industrial Conversion Stage. The results of the exergy analysis of the industrial conversion of rapeseeds, soybeans, and corn kernels into biofuels are presented in Table 1. To convert the crops from 1 ha, with exergy contents of 86.0, 53.8, and 134 GJ, nonrenewable inputs (fuels, electricity, steam and chemicals) of 5.26, 5.72 and 4.96 GJ are required. Next to biofuels, the conversion results in a significant amount of byproducts that can be marketed. The shares of the biofuels in the global output are limited to 57.7 and 29.8% for RME and SME, respectively, due to the high exergy content of the byproducts glycerol and particularly meal. Ethanol shows the highest share in industrial output: 61.4%. With overall outputs of 82.3, 55.0, and 112 GJ for rapeseed, soybean, and kernel conversion, efficiencies of 90.1, 92.4, and 80.6% are obtained, respectively. Note that the auxiliary inputs also require their own industrial production, starting from non-

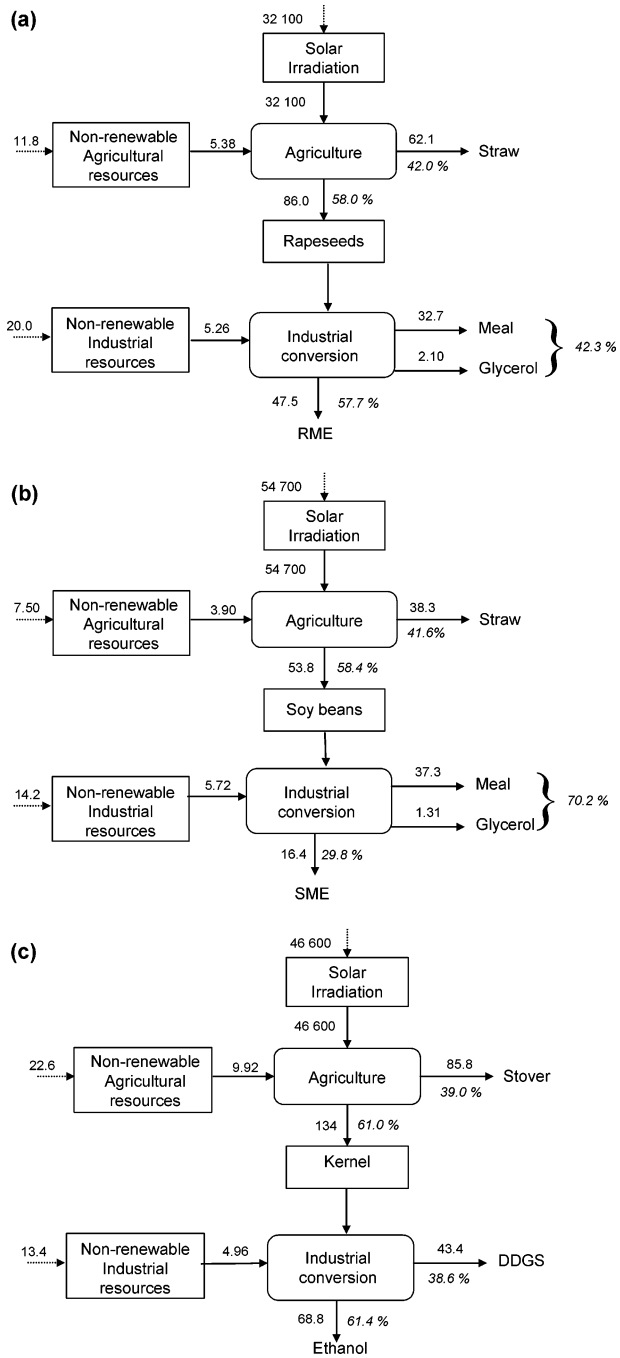

FIGURE 2. Flows of exergy associated with the annual production of biofuels on 1 ha (GJ ha^{-1} yr^{-1}). The flows are related to the overall production, that is, including byproducts. Percentages are the shares in the output of the respective process on an exergy basis. Data at dotted lines are the cumulative exergy extracted out of the natural environment. (a) RME, (b) SME, (c) EtOH.

TABLE 2. Exergy and Cumulative Exergy Inputs Related to the Production of Biofuel from 1 ha of Agricultural Land, with Allocation of Inputs to Biofuel, and Agricultural and Industrial Byproducts

		biofuel	agr. byproducts	ind. byproducts	total
		Exergy Inputs			
RME					
solar irradation	%	58.0 × 57.7 = 33.5	42.0	58.0 × 42.3 = 24.5	100.0
	GJ	10 743	13 482	7875	32 100
nonrenew. agr. resources	%	58.0 × 57.7 = 33.5	42.0	58.0 × 42.3 = 24.5	100.0
	GJ	1.80	2.26	1.32	5.38
nonrenew. ind. resources	%	57.7	0.0	42.3	100.0
	GJ	3.04	0.00	2.22	5.26
SME					
solar irradation	%	58.4 × 29.8 = 17.4	41.6	58.4 × 70.2 = 41.0	100.0
	GJ	9520	22 755	22425	54 700
nonrenew. agr. resources	%	58.4 × 29.8 = 17.4	41.6	58.4 × 70.2 = 41.0	100.0
	GJ	0.7	1.6	1.6	3.9
nonrenew. ind. resources	%	29.8	0.0	70.2	100.0
	GJ	1.70	0.00	4.02	5.72
EtOH					
solar irradation	%	61.0 × 61.4 = 37.5	39.0	61.0 × 38.6 = 23.5	100.0
	GJ	17 454	18 174	10 972	46 600
nonrenew. agr. resources	%	61.0 × 61.4 = 37.5	39.0	61.0 × 38.6 = 23.5	100.0
	GJ	3.72	3.87	2.34	9.92
nonrenew. ind. resources	%	61.4	0.0	38.6	100.0
	GJ	3.0	0.0	1.9	5.0
		Cumulative Exergy Inputs			
RME					
solar irradation	%	58.0 × 57.7 = 33.5	42.0	58.0 × 42.3 = 24.5	100.0
	GJ	10 743	13 482	7875	32 100
nonrenew. agr. resources	%	58.0 × 57.7 = 33.5	42.0	58.0 × 42.3 = 24.5	100.0
	GJ	3.9	5.0	2.9	11.8
nonrenew. ind. resources	%	57.7	0.0	42.3	100.0
	GJ	11.5	0.0	8.5	20.0
SME					
solar irradation	%	58.4 × 29.8 = 17.4	41.6	58.4 × 70.2 = 41.0	100.0
	GJ	9520	22 755	22 425	54 700
nonrenew. agr. resources	%	58.4 × 29.8 = 17.4	41.6	58.4 × 70.2 = 41.0	100.0
	GJ	1.3	3.1	3.1	7.5
nonrenew. ind. resources	%	29.8	0.0	70.2	100.0
	GJ	4.2	0.0	10.0	14.2
EtOH					
solar irradation	%	61.0 × 61.4 = 37.5	39.0	61.0 × 38.6 = 23.5	100.0
	GJ	17 454	18 174	10 972	46 600
nonrenew. agr. resources	%	61.0 × 61.4 = 37.5	39.0	61.0 × 38.6 = 23.5	100.0
	GJ	8.5	8.8	5.3	22.6
nonrenew. ind. resources	%	61.4	0.0	38.6	100.0
	GJ	8.2	0.0	5.2	13.4

renewables. For example, to supply the required 5.26 GJ to process rapeseeds into RME, extraction of 20 GJ of nonrenewables out of the environment has to be taken into account.

Overall Production Chain Assessment. In Figure 2, the overall production chains of RME, SME, and EtOH are represented. This figure allows allocation of renewable and particularly nonrenewable resources to the delivered biofuels and agricultural and industrial byproducts on the basis of exergy. The exergy-based allocation is illustrated in Table 2. Shares of rapeseeds, soybeans, and corn kernels in the agricultural output are 58.0, 58.4, and 61.0%, respectively, allowing the allocation of solar irradiation and nonrenewable agricultural resources to rapeseeds, soybeans, and corn kernel, respectively. Shares of RME, SME, and EtOH in the industrial output are 57.7, 29.8, and 61.4%, respectively, allowing allocation of both the input agricultural product and nonrenewable industrial resources to the biofuel. Overall, this means that the agricultural inputs, to be allocated to the final biofuel in two steps, are limited to 33.5, 17.4, and 37.5%.

It turns out that allocated agricultural and industrial resource inputs for 47.5 GJ of RME are 1.80 and 3.04 GJ, respectively (Table 2), or 4.84 GJ in total. This agricultural–industrial production chain shows a BF_{ex} value of 9.8. If one considers the whole industrial metabolism for this biofuel production, that is, not only the agricultural–industrial biofuel production chain but also the agricultural and industrial resource supply chains, one ends up with nonrenewable resource inputs of 3.9 and 11.5 GJ for the agricultural and industrial stage, respectively, or 15.4 GJ in total. This results in an overall BF_{ex} value of 3.1. In other words, it can be stated that the renewable fraction of RME is limited to 67.6%. Similar calculations can be made for SME and EtOH; the results are represented in Table 3. It turns out that not only for the agricultural production (see above) but also for the overall production chain, corn-based ethanol proves to be the most suitable biofuel in terms of efficiency and renewability. Indeed, it shows an overall industrial metabolism exergy breeding factor (overall BF_{ex}) of 4.1, resulting in the lowest nonrenewable fraction being about one-quarter (24.3%). RME and SME show overall BF_{ex} values of ~3 and a nonrenewable fraction of about one-third.

Shifts in breeding factors may change in the future due to several reasons. First, it is noteworthy to mention that industrial processing of renewable resources is a rapidly developing field (*27*). Implementation of new technologies, for example, conversion of cellulose into ethanol, may result in a better valorization of these resources, eventually resulting in higher breeding factors. Second, when considering the

TABLE 3. Nonrenewable Resources Allocated to the Biofuel (GJ ha^{-1} yr^{-1}) for the Agricultural–Industrial Biofuel Production Chain and for the Overall Industrial Metabolism[a]

	input biofuel production chain	input overall industrial metabolism	biofuel	BF_{ex}	overall BF_{ex}	non-renewable fraction (%)
RME	4.84	15.4	47.5	9.8	3.1	32.4
SME	2.40	5.61	16.4	6.9	2.9	34.2
EtOH	6.72	16.7	68.8	10.2	4.1	24.3

[a] That is, agricultural resource supply chain, industrial resource supply chain, and the biofuel production chain. Exergy breeding factor of the biofuel production chain (BF_{ex}) and of the overall industrial metabolism (overall BF_{ex}) and nonrenewable fraction in the biofuel.

processes under investigation in this study, shift can be realized by substitution of nonrenewable resources. Usage of biofuels in the agricultural machinery can be considered as a first option here. By doing so, breeding factors can shift from 9.8, 6.6, and 10.2 toward 12.1, 9.2, and 18.7 for RME, SME, and EtOH, respectively. Alternatively, substitution of nonrenewables at the industrial stage might realize BF factors up to 26.2, 24.2, and 18.5, respectively. These values for RME and SME show that RME and SME biodiesels currently rely significantly on nonrenewable use in the industrial stage, which is limited to a smaller extent in case of bioethanol.

Supporting Information Available

EtOH, RME, and SME overall experimental data and details. This material is available free of charge via the Internet at http://pubs.acs.org.

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