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An Approach to Determine Missing Life Cycle Inventory Data for Chemicals (RREM)

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Abstract: Chemicals impact the environment. However, life cycle assessments (LCA) of products containing chemicals are often not possible due to a lack of available datasets. Existing methodologies to address this problem have several shortcomings. Therefore, a new approach to model chemicals is introduced to fill dataset gaps in inventory databases. Further data for 60 chemicals are provided. The approach consists of four steps: (i) general research on the chemical and the synthesis processes, (ii) setting up the reaction equations, (iii) researching the required thermal energy, and (iv) modeling of the dataset (RREM). Depending on the obtained data, calculations are carried out or assumptions are applied. The environmental impact of the chemicals is modeled in the LCA software linking to existing datasets. A case study of the chemical octocrylene illustrates the application of RREM. An overview is given of the environmental profile of 60 chemicals modeled based on RREM. The validity of the assumptions and their influences on the results are examined by a sensitivity analysis. By modeling chemicals with the RREM approach, previously unknown environmental impacts of chemicals and products containing them can be determined.

Keywords: life cycle assessment (LCA); life cycle inventory (LCI); data gaps; chemicals; stoichiometry; RREM

Citation: Huber, E.; Bach, V.; Holzapfel, P.; Blizniukova, D.; Finkbeiner, M. An Approach to Determine Missing Life Cycle Inventory Data for Chemicals (RREM). *Sustainability* **2022**, *14*, 3161. https://doi.org/10.3390/su14063161

Academic Editors: Marc A. Rosen and Shervin Hashemi

Received: 21 December 2021
Accepted: 4 March 2022
Published: 8 March 2022

1. Introduction

The environmental impact of products, processes, and services can be determined with the life cycle assessment (LCA) methodology according to ISO 14040/44 [1,2]. One result of an LCA is to identify a product's contribution to various impact categories such as climate change, eutrophication, and acidification [3]. For this, the entire product life cycle must be considered, including the extraction of raw materials, the manufacturing of the product, its usage, recycling, etc.

An essential part of every LCA is the life cycle inventory (LCI) [4]. The LCI includes data collection where all input and output flows are collected for all processes within the system. In this procedure, material and energy flows are traced back to the extraction of resources from the environment and to emissions into the environmental compartments such as soil, air, and freshwater. The determination of these so-called elementary flows requires a large amount of data, which can be derived from several sources.

Primary data sources often provide the best data quality, since they relate specifically to the product under investigation [5]. These include recordings from process control systems and direct measurements of, for example, emissions, permit documents, and waste balances [6]. However, primary data are often not available, especially for background systems (up- and downstream processes) [7,8]. When primary data are missing, different sources are used to obtain secondary data. Possible data sources are average industry data, results of other LCA case studies, and data from generic databases such as GaBi [9] or Ecoinvent [10]. Further, proxy data [5] can be used to enable qualified estimations of environmental impacts, which are based on similar value chains and production processes.

In addition, data from technical literature and process simulation can be applied for modeling. Regardless of which data source is used, it must be in line with the defined goal and scope.

Existing LCI databases include many substances and processes. The GaBi and Ecoinvent databases, taken together, contain over 30,000 datasets [9,11]. However, there are still data gaps, for example, for the chemicals ascorbyl palmitate, tocopheryl acetate, and zinc carbonate. These gaps need to be filled in a daily LCA routine, which requires the collection of a large amount of information. This is often time-consuming, costly, and in some cases not possible at all [12,13]. Therefore, other sources of secondary data must be applied to model chemicals.

Chemicals have significant environmental relevance because they can enter the environmental compartments via numerous pathways, and therefore, might also pose a risk to human health and ecosystems [14–16]. Their production is also of environmental importance, as the chemical sector is one of the most energy-intensive industries [17] and requires more than 20 million tons of raw materials in Germany alone [18]. Approximately 20,000 chemicals are used commercially in Europe, over 23,000 are registered at the European Chemical Agency and to date, and more than 80 million have been described in the scientific literature [14,19].

Existing methods provide different approaches to fill data gaps for chemicals. Milà i Canals et al. [20] examined proxy datasets (scaled proxies, direct proxies, and averaged proxies) and data exploration in terms of variability and uncertainty when used as surrogate data for biogenic products. Subramanian and Golden [13] analyzed the expert elicitation methodology and Meron et al. [21] developed the "selection proxy" methodology.

Wernet et al. [22] introduced a model for closing data gaps for chemicals by deriving information about required resources and energy from molecular structure. This avoids a data-intensive process-based model and helps when extensive analysis is not possible due to a lack of data or resources. Nevertheless, process-based models seem to provide more accurate assessments, which is why Wernet et al. [23] recommended a staggered combination of molecular structure- and process-based models due to the easily achievable results, which still have sufficient quality. However, there is still an opportunity to improve the quality by taking a reaction-specific view and including other environmental impact categories.

A method based on stoichiometric equations was developed by Hischier et al. [24] for cases of poor data availability. With this method, data gaps can be filled using the reaction equation and assumptions on yield, energy and water consumption, emissions to air and water, waste, and transport. Nevertheless, the data obtained with this method can only be used if the chemical under consideration is not a main input of the analyzed product system, as they are associated with many uncertainties.

Geisler et al. [25] developed an input–output model based on stoichiometry. To apply their method, molar masses of all compounds, stoichiometric coefficients, and basic information about the reaction medium must be known. Missing data on, for example, emissions, solvent masses, and recycling are estimated with default values and evaluated with scenario analyses. The values used for best-case and worst-case scenarios partly cover a wide range, for example, the estimated values for the yield are 95% or 0%. There are no estimates for catalysts, although these can make a considerable contribution to the LCA of a chemical.

In summary, existing methodologies have the following weaknesses:

- Results can be obtained only for a few impact categories which limits the analysis and interpretation (Wernet et al. [22]).
- Important specific reaction parameters are not considered which can lead to an underestimation of the environmental impacts (Wernet et al. [23]).
- Due to uncertainties in the results, the method is only applicable when chemicals have a small contribution in the LCA (Hischier et al. [24]).

- Synthesis components, such as catalysts, with potentially high environmental impacts are not considered, leading to an underestimation of the environmental impacts (Geisler et al. [25]).

In this paper, we aim to address the gaps in LCI databases and existing methodologies by introducing a new and practical approach to fill missing LCI datasets for chemicals, referred to as RREM (research, reaction, energy, and modeling). The significance of RREM is that it allows a more precise estimation of environmental impacts of chemicals. RREM enables the creation of datasets, which are the basis to determine results of all impact categories available in the used software.

Further, we aim to deliver results achieved with RREM. We provide the environmental profile of 60 chemicals, containing global warming potential for 100 years excluding biogenic carbon (GWP100), acidification potential (AP), and eutrophication potential (EP). Additionally, a detailed case study is presented to illustrate the application of RREM.

Moreover, the following compiled background data needed for modeling are provided in Supplementary Materials:

- Excel® file for 60 chemicals with the summarized data needed for modeling datasets;
- Excel® file with an overview of the heat capacities and densities required for the calculations.

In the following, RREM is described (Section 2) and illustrated by a case study of octocrylene (Section 3). The environmental profile of the 60 chemicals determined with RREM is presented (Section 4). Weaknesses are discussed and key assumptions are examined in a sensitivity analysis (Section 5). Finally, conclusions are drawn (Section 6).

2. RREM Approach

The RREM approach is applicable for chemicals when no dataset and no suitable proxy are available in databases. It enables the modeling of chemicals to fill gaps in LCI data. RREM consists of the following four steps (see Figure 1):

1. Research on the chemical and its synthesis process;
2. Setting up of the reaction equations and checking data availability with existing databases;
3. Research on the thermal energy demand;
4. Modeling of the dataset and connecting to existing datasets.

Sections 2.1–2.4 describe the procedure and assumptions of RREM in detail.

2.1. Step 1: Research on the Chemical and Its Synthesis Process

General information is gathered in the first step by desk research. This includes the chemical formula, the IUPAC name, other names for the chemical, the CAS registry number, and the molar mass.

Since literature data usually do not contain information about environmental effects, the environmental impacts of the chemical are quantified by modeling its production. Therefore, synthesis data, such as descriptions and schemes of the performed reactions are collected from the literature, preferably publicly available patents.

2.2. Step 2: Setting Up of the Reaction Equations and Checking Data Availability with Existing Databases

In the second step, reaction equations are set up with the collected information (see Equation (1)). Data on the synthesis reaction are compiled and calculated stoichiometrically. These include the chemical name and formula, reaction coefficient, molar mass, chemical amount, and mass of the necessary reactants (R) and formed products (P). The coefficients and potential by-products can be derived by balancing the reaction equation stoichiometrically.

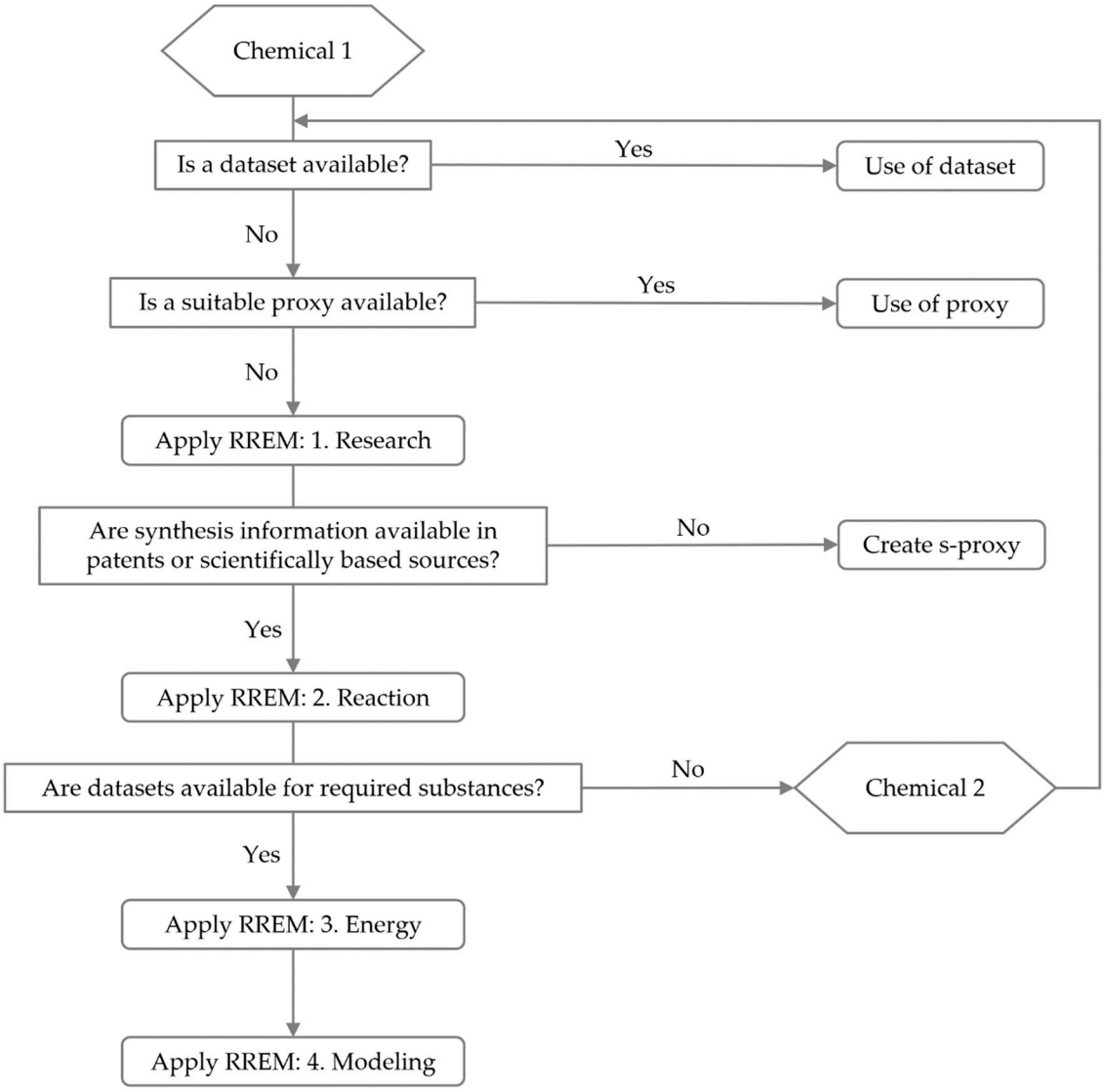

Figure 1. Issues and decision making before and during the application of RREM. Hexagons represent chemicals; rectangles represent questions that arise when applying RREM; rectangles with rounded corners represent actions to be taken.

In addition, the quantity of reactants that do not react is calculated by subtracting the chemical amount of the product from the chemical amount of the reactant. It is assumed that 100% of the unreacted reactants are returned to the reaction. This assumption applies unless other information is available:

$$R1 + R2 \rightarrow P1 + P2 + \text{Unreactant } R1 + \text{Unreacted } R2 \quad (1)$$

In the case that data are missing, the required quantities of reactants can be determined according to the following procedure:

The molar mass M (g·mole^{-1}) can be calculated according to Equation (2) using the chemical amount n (mole) or the mass m (g). The mass can be calculated based on volume V (ml) and density ρ (g·ml^{-1}) as:

$$M = \frac{m}{n} \quad (2)$$

The chemical amount of the product n_p is calculated via the yield y and the chemical amount of the limiting reactant n_l (see Equation (3)) as:

$$n_p = y * n_l \tag{3}$$

However, data on quantities may be missing. In the worst case, no information on the synthesis process can be obtained, neither on the substances nor on their quantities. In this case, the following assumptions are made. These assumptions apply unless other information is provided in the considered sources.

If data on the quantity of products are missing, it is assumed that the yield is 70% based on the limiting reactant. This value is based on the experience from the application of RREM.

If information on the quantity of reactants is missing, it can be assumed that the stoichiometric coefficient corresponds to the chemical amount of the reactant. A coefficient of 1 corresponds to a chemical amount of 1 mole. In the case that the chemical amount is given for one reactant and only the quantities of further reactants are missing, the given chemical amount can be transferred according to the coefficients.

Data on catalysts and solvents required for the synthesis are also collected. These include the chemical name and formula, molar mass, and quantity. Data collection is done analogously to the approach for the reactants. It is not specifically defined whether the substance is a catalyst or a solvent, because it is only relevant that it is not consumed during the reaction. If data on required catalysts and solvents are available, but quantity data are missing, estimates are made based on the available reaction quantity and similar reactions.

In addition, information on the recycling rate and disposal pathway is collected. When specific data are not available, assumptions are defined. It is assumed that a defined fraction of the catalysts and solvents is returned to the reaction (see explanation of recycling rates in the following paragraphs). As for the other assumptions, the recycling rates apply unless other information is available in the considered sources. The remaining fraction is disposed, according to the substance properties as hazardous waste with or without carbon (C), wastewater, organic waste, or residual waste. Here, a conservative approach is chosen which results in an overestimation of the environmental burdens to compensate for weaknesses of the assumptions. Waste is usually classified as hazardous unless it is certain that the substance is non-hazardous. Wastewater is assumed to be disposed of via a wastewater treatment plant, instead of evaporation.

For catalysts and solvents, a general recycling rate of 50% is assumed, according to existing literature (e.g., [26–29]). For solvents, the recycling rate can vary between 70% and 100% [26,27]. For catalysts, the recycling rate is particularly dependent on their properties [28,29]. For homogeneous catalysts, recycling is very difficult or not possible at all, leading to a recycling rate of 0%. Whether or not a catalyst is homogeneous depends on the reaction system and cannot be generalized. The categorization of a substance as a solvent, heterogenous or homogeneous catalyst is challenging, because it depends on the reaction system. As there is no separation made in RREM between solvents and catalysts, 50% recycling rate is assumed.

For certain substances used as catalysts and solvents, a distinct consideration is needed, and therefore, considered in RREM. This applies to silica gel and transition metals in pure form. Silica gel can be recycled 10 times without reducing the separation performance [30], leading to a recycling rate of 90%. Transition metals used as catalysts are expensive and environmentally harmful [31]. Therefore, the goal is a high recirculation rate. The recycling of platinum group metals ((PGMs) belonging to the group of transition metals) is very efficient in industrial processes [32]. Recycling rates of over 90%, and in some cases over 99%, are achieved for PGMs [33]. Moreover, PGMs can be used as catalysts for several successive years [34]. This leads to the assumption of a recycling rate of 99% for pure PGMs. Since no clear data could be found for other transition metals, this assumption was adopted for them (when they are used in their pure forms) as well to provide a value for the application of RREM.

The assumptions on the recirculation of reactants, catalysts, and solvents are based on key aspects of the chemical industry. Resource efficiency and circular economy are already relevant and will become even more important to save costs as well as improve on environmental performance [35]. This makes the reuse and recycling of raw materials necessary. Therefore, it is assumed that the components of the reaction are kept in circulation for as long as they can be used without loss of performance. These assumptions are further addressed in the sensitivity analysis (see Section 5).

For all inputs, i.e., reactants, catalysts, and solvents, information is collected regarding availability of datasets or suitable proxies in existing databases. If datasets are available, production processes of the considered region are preferably selected. If there are no suitable datasets and no proxies available, RREM (see Step 1–4) is applied analogously.

In the case that no synthesis information is available, neither patents nor other scientifically based sources, a special proxy, hereafter called s-proxy, is derived. The s-proxy is modeled based on the chemical structure and the molar mass of the considered substance. Substances that, in combination, have approximately the same chemical structure and molar mass as the considered substance are assumed to be reactants. Datasets for the assumed reactants can, then, by selected in existing LCI databases. Catalysts, solvents, and waste are not included, and it is assumed that reactants are completely converted during reactions. This tends to underestimate the environmental impact. To compensate for it, the input quantities are multiplied by a factor of two, the so-called safety factor. The influence of the safety factor is further examined in the sensitivity analysis (see Section 5). Further, possible by-products are not considered in s-proxies.

2.3. Step 3: Research on the Thermal Energy Demand

In the third step, the thermal energy required for synthesis is determined. It cannot be derived from stoichiometry but its provisioning contributes to the environmental impacts [36]. RREM focuses on this required thermal energy only, disregarding energy for other process components such as machinery.

If information on temperature is available, the required thermal energy can be calculated as follows: First, data on heat capacities of the reactants, catalysts, and solvents are gathered. They are converted from the molar heat capacities c_v (MJ·mole^{-1}·K^{-1}) into the specific heat capacities c_p (MJ·kg^{-1}·K^{-1}) via the molar mass M (g·mole^{-1}), according to Equation (4) as:

$$c_p = \frac{c_v}{M * 1000} \tag{4}$$

The average specific heat capacity $\varnothing c_p$ is calculated based on the specific heat capacities c_p and the masses m_k of the substances used in the synthesis reaction (see Equation (5)) as:

$$\varnothing c_p = \frac{\sum_{k=1}^{n}(c_{pk} * m_k)}{\sum_{k=1}^{n} m_k} \tag{5}$$

Thus, the thermal energy demand of the reaction Q_r (MJ) can be calculated by multiplying the average heat capacity $\varnothing c_p$, the reaction mass m_r, and the temperature change ΔT (K), according to Equation (6) as:

$$Q_r = \varnothing c_p * m_r * \Delta T \tag{6}$$

If the starting temperature is not given, a typical room temperature of 20 °C is assumed. In the case that the reaction requires cooling down to below 20 °C, this is considered when calculating the temperature difference, as well as the temperature changes during the reaction.

When temperature data are missing, or an s-proxy is created, it is assumed that the synthesis of 1 kg product requires 1.8 MJ of thermal energy. This value is based on the experience from the application of RREM and is further addressed in the sensitivity analysis (see Section 5).

2.4. Step 4: Modeling of the Dataset and Connecting to Existing Datasets

In the fourth step, the necessary data to model the chemical are summarized to prepare the LCI data. These include the input and output masses, which were researched and calculated in Step 2, and the calculated thermal energy from Step 3. Data on disposal, recycling, and databases are included as well.

If relevant by-products are formed, the allocation factor is calculated based on the mass according to ISO 14044 [2]. Water as a by-product is not considered in the allocation since it is assumed that it cannot be used for further synthesis processes without treatment.

The compiled data are used to create the dataset of the considered chemical using LCA software, for example, SimaPro [37] and GaBi [38]. This is done by connecting material and energy flows in the usual manner. The connection to existing LCI datasets is an essential part of RREM, as it provides the assignment of elementary flows and environmental impact to the considered chemical.

Since RREM is generic, the LCA software and database are not predefined. In Sections 3–5, datasets are modeled in GaBi (Version 10) using the Sphera (Version 2021.2) and Ecoinvent (Ecoinvent 3.5) databases. The impacts are determined using the CML 2001 method [39].

3. Case Study: Octocrylene

This section describes the application of the RREM approach using the case study of octocrylene for which no dataset or proxy is available in common LCI databases. It is chosen because the steps and assumptions of RREM can be effectively explained. The individual data collection, compilation, and calculation steps necessary to create a dataset for octocrylene are described in the following subsections. Based on the datasets modeled, the GWP100 is determined.

First, the application of RREM for octocrylene is covered (Section 3.1). This is followed by the application for three chemicals required for the synthesis of octocrylene and for their reactants (Sections 3.2–3.6). Figure 2 provides an overview of the chemicals modeled with RREM for the case study. For the other needed chemicals, RREM does not have to be applied, because a direct connection to existing datasets in GaBi or Ecoinvent is possible (see corresponding Excel® files).

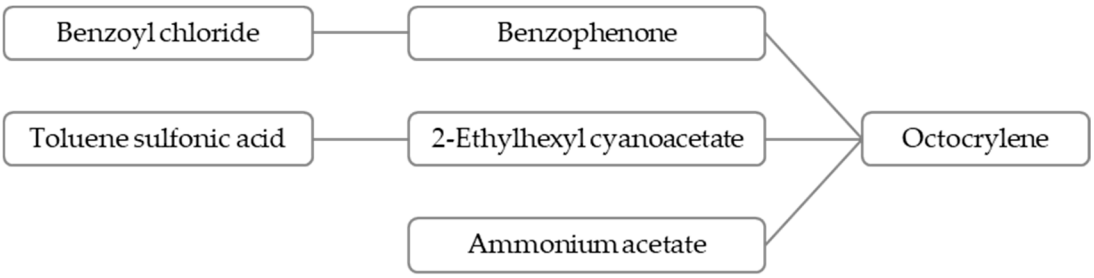

Figure 2. Overview of chemicals modeled using RREM for the octocrylene case study.

3.1. Octocrylene

Octocrylene is a synthetically produced UV filter, typically used in sunscreens [40]. General information is summarized below:

Chemical formula	$C_{24}H_{27}NO_2$
IUPAC name	2-Ethylhexyl-2-cyano-3,3-diphenylprop-2-enoate
Another name	Uvinul N539
CAS registry number	6197-30-4
Molar mass	361.50 g·mole^{-1}

Information on the synthesis is obtained from patent no. US5451694A [41], which describes the synthesis of octocrylene ($C_{24}H_{27}NO_2$): benzophenone ($C_{13}H_{10}O$) and 2-ethylhexyl cyanoacetate ($C_{11}H_{19}NO_2$) are needed as reactants (see Equation (7)); propionic

acid, ammonium acetate, and water are listed as catalysts/solvents required for the reaction. The only defined by-product is water (H_2O), which is not considered in the allocation, leading to an allocation factor of 1. The mass balance reveals that 28.73 g of an unknown substance is produced. Since the substance cannot be determined and the reactants contain carbon, it is categorized as a carbon-containing hazardous waste. Equation (7) is expressed as:

$$C_{13}H_{10}O + C_{11}H_{19}NO_2 \rightarrow C_{24}H_{27}NO_2 + H_2 \quad (7)$$

When checking for which chemicals, LCI datasets or appropriate proxies are available, preferentially production processes within the EU, it is noted that datasets are lacking for three chemicals. Thus, RREM also has to be applied to benzophenone, 2-ethylhexyl cyanoacetate, and ammonium acetate (see Sections 3.2–3.4) to complete the modeling of octocrylene.

To determine the thermal energy demand, the specific heat capacities of the substances are investigated. The heat capacities for 2-ethylhexyl cyanoacetate and ammonium acetate are not available. Thus, the known data on benzophenone and propionic acid are used to calculate the average heat capacity. The initial temperature of the entire reaction is not given in the patent; therefore, the assumption of 20 °C is applied in Equation (6). Since water is used for washing after the mixture was heated up, it is not accounted for.

The dataset of octocrylene modeled in GaBi is shown in Figure 3. Input processes are the processes for the reactants, catalysts, solvents, and energy. The inputs benzophenone, 2-ethylhexyl cyanoacetate, and ammonium acetate are processes modeled based on RREM (see Section 2), since no LCI datasets are available. The specified outputs are reused or recycled, as described in Section 2.2. In the case of catalysts and solvents, the non-recirculated fraction is passed on to the corresponding waste processes. The same applies to the amount of the unspecified substance.

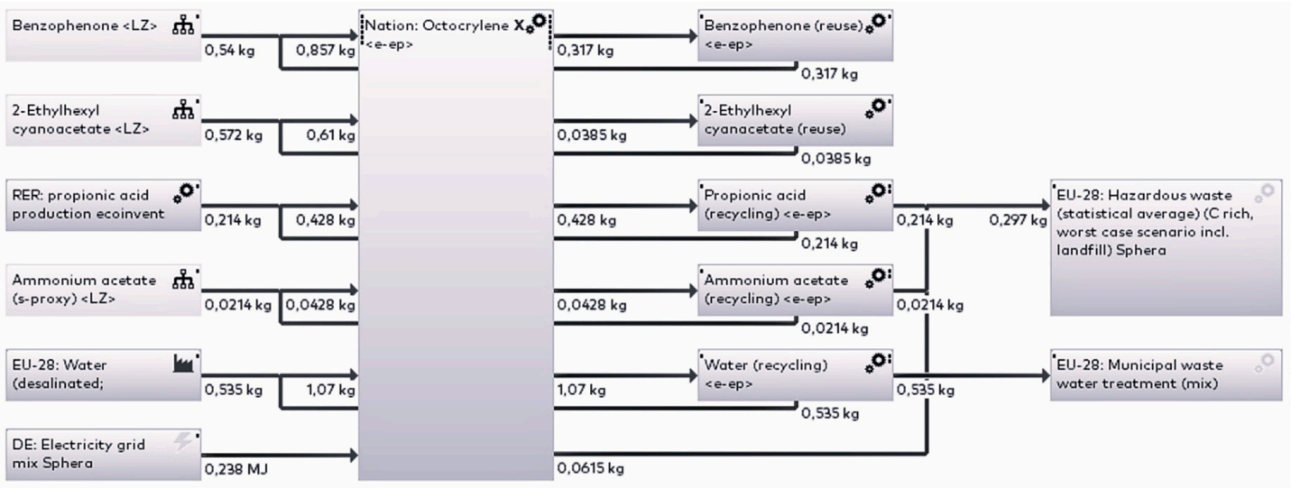

Figure 3. Model of octocrylene carried out in GaBi.

Octocrylene has a GWP100 of 7.09 kg CO_2eq per kg (see Figure 4). Approximately 90% of the impact can be attributed to the two reactants benzophenone and 2-ethylhexyl cyanoacetate. Benzophenone causes 36% and 2-ethylhexyl cyanoacetate accounts for more than half of octocrylene's GWP100. The remaining emissions are caused by propionic acid, the disposal of hazardous waste, ammonium acetate, and energy.

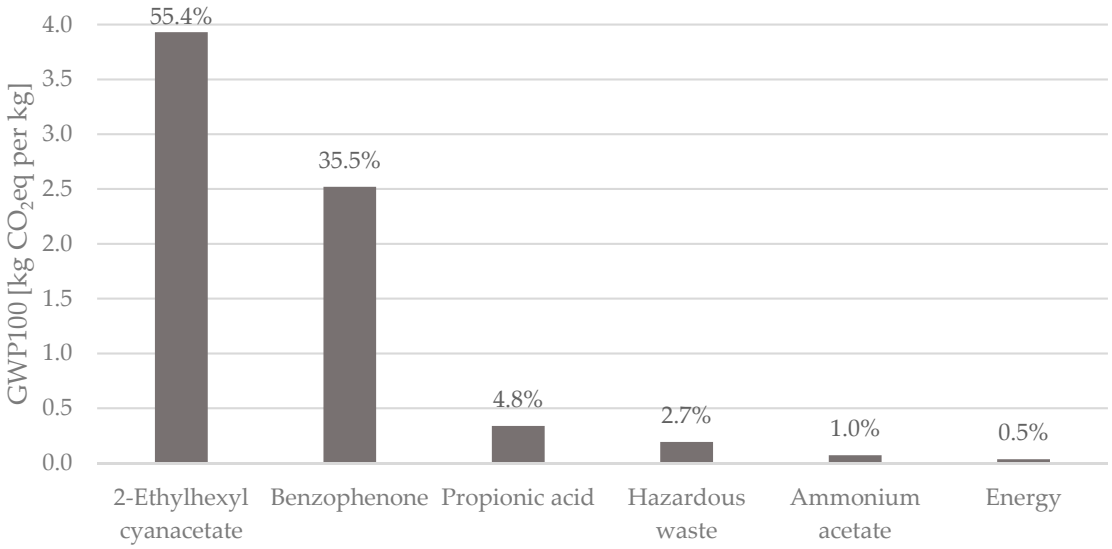

Figure 4. Contribution of the components to the GWP100 of 1 kg octocrylene.

3.2. Benzophenone

Benzophenone is one of the reactants required for octocrylene synthesis. Since no LCI dataset or appropriate proxy is found, it is necessary to obtain the data using RREM.

Benzophenone ($C_{13}H_{10}O$) can be synthesized by reaction of benzene (C_6H_6) and benzoyl chloride (C_7H_5ClO) in the presence of aluminum chloride (see Equation (8)) [42]. During the synthesis of benzophenone, hydrochloric acid (HCl) is formed as a by-product, resulting in an allocation factor of 0.83. Equation (8) is expressed as:

$$C_6H_6 + C_7H_5ClO \rightarrow C_{13}H_{10}O + HCl \quad (8)$$

Since only the necessary volume is given for benzene, the mass is calculated via its density. For benzoyl chloride, no LCI dataset or suitable proxy is available. Thus, it must be modeled by applying RREM (see Section 3.5). Due to missing information on the reaction temperature, a demand of 1.8 MJ thermal energy per 1 kg benzophenone is applied, as described in Section 2.3.

The GWP100 of benzophenone is 2.52 kg CO_2eq per kg octocrylene. More than 80% are caused by aluminum chloride and benzoyl chloride. Benzene, thermal energy, and the disposal of hazardous waste accounts for the remaining 16%.

3.3. 2-Ethylhexyl Cyanoacetate

2-Ethylhexyl cyanoacetate is one reactant required for octocrylene synthesis. RREM is applied, since neither an LCI dataset nor a suitable proxy is available. Patent no. TWI422560B provides information on the synthesis of 2-ethylhexyl cyanoacetate ($C_{11}H_{19}NO_2$) (see Equation (9)) [43]. For the synthesis, cyanoacetic acid ($C_3H_3NO_2$) and 2-ethylhexanol ($C_8H_{18}O$) are reactants, toluene sulfonic acid and water are used as catalysts/solvents. The reaction reaches a yield of 92% based on cyanoacetic acid (see [43]). Based on the yield, the quantity of the products can be calculated. The only by-product is water, leading to an allocation factor of 1. Equation (9) is expressed as:

$$C_3H_3NO_2 + C_8H_{18}O \rightarrow C_{11}H_{19}NO_2 + H_2O \quad (9)$$

For the reactant toluenesulfonic acid, no dataset is available; thus, RREM is applied (see Section 3.6). For 2-ethylhexanol, no LCI dataset is available either, but a dataset for cyclohexanol is available, which is applied as an s-proxy.

The specific heat capacities of all substances were found to calculate the required thermal energy. The starting temperature is assumed to be 20 °C. The patent mentions that

the reaction mixture is first heated to 130 °C, and then again from 90 °C to 130 °C. This results in a total temperature difference of 150 °C.

2-Ethylhexyl cyanoacetate has a GWP100 of 3.93 kg CO_2eq per kg octocrylene, which originates almost entirely from its reactants. The contribution of cyclohexanol is 69%, and that of cyanoacetic acid is 30%. Thermal energy causes only 1%.

3.4. Ammonium Acetate (S-Proxy)

Ammonium acetate is used as a catalyst/solvent for the synthesis of octocrylene. Detailed information including the description of the synthesis is lacking for ammonium acetate. Since only a minimum of information on the reaction is available, an s-proxy is created using existing databases.

Based on the chemical formula and molar mass, ammonium acetate can be composed of acetic acid and ammonia. The molar mass of ammonium acetate ($C_2H_7NO_2$) is 77.08 g·mole^{-1}, that of acetic acid ($C_2H_4O_2$) is 60.05 g·mole^{-1}, and that of ammonia (NH_3) is 17.03 g·mole^{-1}. Therefore, one molecule of acetic acid and one of ammonia can form one molecule of ammonium acetate (see Equation (10)) as:

$$C_2H_4O_2 + NH_3 \rightarrow C_2H_7NO_2 \quad (10)$$

Based on the reaction coefficients, one mole is assumed as input for both chemicals. The established assumption regarding a thermal energy of 1.8 MJ per 1 kg of product is applied, considering the safety factor.

3.5. Benzoyl Chloride

Benzoyl chloride (C_7H_5ClO) is required as a reactant for modeling benzophenone. It can be synthesized by the reaction of benzaldehyde (C_7H_6O) and chlorine (Cl_2) (see Equation (11)) [44]. Hydrochloric acid is produced as a by-product, which is considered in the allocation, resulting in an allocation factor of 0.79. Equation (11) is expressed as:

$$C_7H_6O + Cl_2 \rightarrow C_7H_5ClO + HCl \quad (11)$$

LCI datasets are available for both chemicals. In the reaction description, the amount of chlorine is missing. Since benzaldehyde and chlorine both have a reaction coefficient of 1, the chemical amount of benzaldehyde is also applied for chlorine. As the quantity of product is not specified, a yield of 70% is assumed. No information on the reaction temperature is provided, so a thermal energy requirement of 1.8 MJ per 1 kg benzoyl chloride is assumed.

Based on the model, benzoyl chloride has a GWP100 of 1.03 kg CO_2eq per kg octocrylene. The input benzaldehyde has the largest contribution with 82%. The contribution of chlorine is 13%.

3.6. Toluenesulfonic Acid (S-Proxy)

No detailed descriptions of the production of toluenesulfonic acid can be found. Therefore, an s-proxy is created.

One molecule of toluene (C_7H_8) and one of sulfuric acid (H_2SO_4) can form one molecule of toluenesulfonic acid ($C_7H_8O_3S$) (see Equation (12)). Here, water would also be produced, but this is not considered, and 1 mole of each reactant is needed since the reaction coefficients are 1. Due to the lack of information, the demand for thermal energy is set to 1.8 MJ per 1 kg of product. Equation (12) is expressed as:

$$C_7H_8 + H_2SO_4 \rightarrow C_7H_8O_3S\ (+H_2O) \quad (12)$$

4. Environmental Profiles

Overall, 60 chemicals are modeled in GaBi using RREM. Thus, their environmental profiles can be determined, which are presented in this section. The environmental profiles

contain GWP100, AP, and EP determined per 1 kg of chemical according to the CML 2001 method. RREM can also be used to determine results of all other impact categories, which are available in the software.

The environmental profiles are summarized in Table 1. For each listed chemical, the Excel® file with the summarized data can be found in the Supplementary Materials.

Table 1. Environmental profiles of chemicals modeled using RREM.

Chemical	GWP100 (kg CO_2eq)	AP (kg SO_2eq)	EP (kg PO_4^{3-}eq)
1-Naphthol-4-sulfonic acid	17.10	0.0825	0.1589
2-(4-(Diethylamino)-2-hydroxybenzoyl)-benzoic acid	10.20	0.0341	0.0140
2,4-Bis-(2,4-dihydroxy-phenyl)-6-(4-methoxyphenyl)-1,3,5-triazine	25.70	0.0797	0.0300
2-Ethylhexyl 4-aminobenzoate	8.26	0.1660	0.0077
2-Ethylhexyl bromide (s-proxy)	3.77	0.0138	0.0047
2-Ethylhexyl cyanoacetate	6.87	0.0280	0.0227
3-Diethylaminophenol	9.17	0.0354	0.0196
4-((4,6-Dichloro-1,3,5-triazin-2-yl)-amino)-benzoic acid	21.60	0.0468	0.0233
4-(4,6-Bis(4-((2-ethylhexyloxy)-carbonyl)-phenylamine)-1,3,5-triazin-2-ylamino)-benzoyl chloride	14.20	0.1160	0.0160
4-(4,6-Bis(4-((2-ethylhexyloxy)-carbonyl)-phenylamine)-1,3,5-triazin-2-ylamino)-benzoic acid	16.00	0.1310	0.0177
6-(4-Methoxyphenyl)-2,4-dichlor-1,3,5-triazine	22.20	0.0754	0.0351
Alpha isomethyl ionone	164.00	6.0400	0.0943
Ammonium acetate (s-proxy)	3.31	0.0029	0.0005
Ascorbyl palmitate	9.07	0.0515	0.0439
Benzophenone	4.66	0.0195	0.0101
Benzoyl chloride	2.96	0.0106	0.0050
Benzyl benzoate (s-proxy)	6.12	0.0187	0.0091
Bis-ethylhexyloxyphenol methoxyphenyl triazine	31.10	0.1330	0.0367
Caprylhydroxamic acid	3.42	0.0635	0.0221
Caprylyl glycol	5.80	0.0088	0.0021
Carmosine	2.57	0.0147	0.0170
Cetearyl isononanoate	2.46	0.0053	0.0011
Citral	224.00	8.6000	0.1270
Dibutyl adipate	4.97	0.0109	0.0033
Diethylamino hydroxybenzoyl hexyl benzoate	12.80	0.0442	0.0151
Diethylhexyl butamido triazone	13.30	0.1060	0.0146
Ethyl p-aminobenzoate	11.40	0.2844	0.0050
Ethylhexyl stearate	2.89	0.0206	0.0147
Iminodiacetonitrile	2.22	0.0028	0.0009
Isoprenol	1.99	0.0028	0.0003
Levulinic acid	2.17	0.0107	0.0044
Methyl caprylate	0.85	0.0087	0.0099
Naphthionic acid	3.17	0.0295	0.0126
Nitrobenzoic acid (s-proxy)	3.02	0.0038	0.0005
Octocrylene	7.09	0.0285	0.0193
Octyldodecanol	4.12	0.0365	0.0522
Palmitoyl chloride	1.20	0.0194	0.0089
PEG-40 hydrogenated castor oil (s-proxy)	2.95	0.0190	0.0114
Phenoxyethanol	1.86	0.0029	0.0005
Prenal	2.26	0.0055	0.0004
Prenol	394.00	15.2000	0.2240
Sodium acetate (s-proxy)	3.30	0.0042	0.0008
Sodium benzoate	1.78	0.0022	0.0003
Sodium levulinate (s-proxy)	4.55	0.0198	0.0077
Sodium naphthionate (s-proxy)	6.45	0.0551	0.0231

Table 1. *Cont.*

Chemical	GWP100 (kg CO_2eq)	AP (kg SO_2eq)	EP (kg PO_4^{3-} eq)
Sodium naphthol sulfonate	13.80	0.0659	0.1260
Tert-butylamine (s-proxy)	2.52	0.0067	0.0006
Tocopherol	10.50	0.0309	0.0189
Tocopheryl acetate	36.60	0.1207	0.0681
Toluenesulfonic acid (s-proxy)	1.52	0.0095	0.0003
Tridecane	3.87	0.0341	0.0498
Triethyl citrate (s-proxy)	3.22	0.0068	0.0025
Trimethylhydroquinone	23.00	0.0530	0.0254
Trimethylhydroquinone acetate (s-proxy)	40.20	0.0994	0.0599
Trisodium dicarboxymethyl alaninate	1.41	0.0022	0.0005
Undecane	3.90	0.0341	0.0497
Urotropine (s-proxy)	3.80	0.0027	0.0012
Zinc carbonate	3.65	0.0104	0.0014
Zinc chloride (s-proxy)	2.70	0.0091	0.0008
Zinc lactate	4.31	0.0146	0.0067

Three chemicals have noticeable GWP100 results: prenol, citral, and alpha isomethyl ionone. The high GWP100 results, which are above average in the listed chemicals, is due to the use of palladium. Palladium is required as a catalyst in the synthesis of prenol, which is needed in the synthesis of citral [45]. Citral is required in the synthesis of alpha isomethyl ionone [46].

5. Discussion

As shown with the case study and the environmental profiles, RREM can be used to fill LCI data gaps and to provide impact assessment results, for example, on GWP100, AP, or EP. However, it must be considered that RREM has some weaknesses, leading to inaccuracies.

The overall quality of the result is highly dependent on the quality of the available sources. Possibilities to fill information gaps with publicly available data are limited. Synthesis reactions in industrial processes may differ from reactions described in the sources that are carried out in laboratories.

RREM does not consider process components, for example, used equipment and machinery, transportation, and water required for cleaning, as well as energy needed for powering machinery. Neglecting these processes and components might lead to an underestimation of the environmental impacts. However, these components are used to produce a high amount of chemicals. Thus, only a small part of their environmental impact would be allocated to the chemical for which RREM is applied.

The assumptions used when information is missing lead to weaknesses as well. The assumed values for yield and thermal energy are based on data from chemicals modeled with RREM. However, other data can also be used as a basis, such as company-specific primary data. Since these are difficult to obtain, substance- or process-specific average values could also be derived to define assumption values. To determine the extent of uncertainty due to the assumptions and their influence on the GWP100, a sensitivity analysis is carried out for the following assumptions:

- Recycling of catalysts and solvents;
- Reuse of reactants;
- Required thermal energy;
- Safety factor for s-proxies.

For each of these assumptions, two scenarios are considered, resulting in a total of eight scenarios. These are illustrated in Figure 5. The reference results are presented in the case study in Section 3.

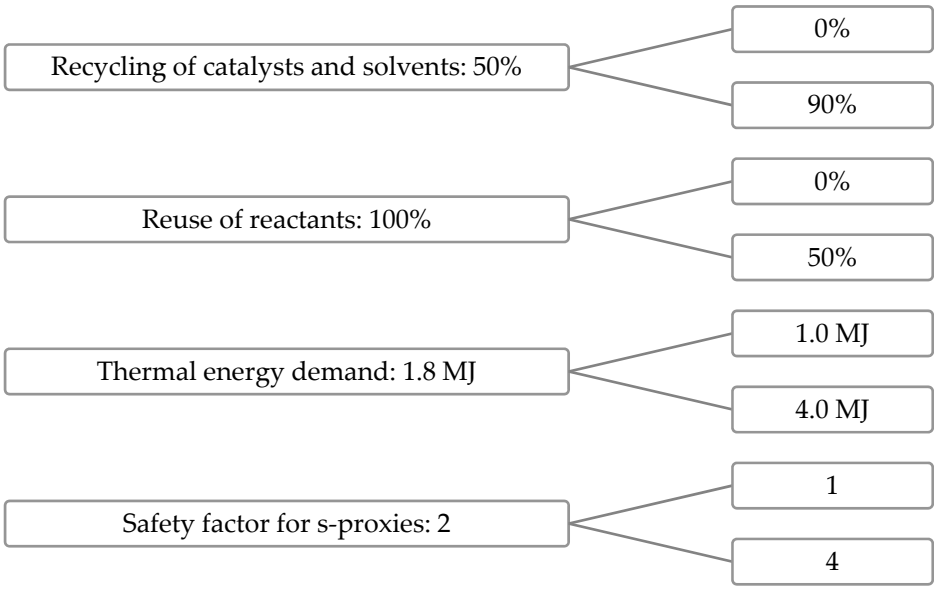

Figure 5. Scenarios considered in the sensitivity analysis to examine the influence of the assumptions.

First, it is necessary to determine how a change in recycling rate for catalysts and solvents affects the result. Thus, a recycling rate of 0% is examined as the worst-case scenario and a recycling rate of 90% as the best-case scenario. In the worst-case scenario, the GWP100 of octocrylene increases by 24.1%, to 8.80 kg CO_2eq per kg (see Figure 6). This is mainly caused by the 45.6% increase in the GWP100 of benzophenone, as this is driven by aluminum chloride. Although the GWP100 of toluenesulfonic acid doubles, it does not affect the overall result substantially (less than 1%) because only a very small amount is used. In the best-case scenario, the GWP100 of octocrylene decreases by 19.5%, to 5.71 kg CO_2eq per kg (see Figure 6). Again, this is mainly due to the change in GWP100 of benzophenone, which decreases by 36.9%.

Therefore, the assumption on the recycling of catalysts and solvents can change the GWP100 of octocrylene by up to a quarter. The influence might also be greater in cases where GWP100 is dominated by catalysts and solvents. To determine which recycling rate is most realistic, each synthesis process would have to be studied in detail. However, this would no longer meet the purpose of RREM, which is to enable a practical way to derive the environmental impact. Nevertheless, the assumption of 50% recycling should be further investigated, especially if catalysts and solvents contribute substantially to the products total GWP100.

Next, the assumption on the reuse of unreacted reactants is investigated. The first scenario of the analysis considers the case of no reuse and the second scenario considers a reuse rate of 50%. In the scenario without reuse, the result changes substantially (see Figure 6). With 23.7 kg CO_2eq per kg, the GWP100 of octocrylene is 3.3 times higher as compared with the reference scenario (+234.3%). This is mainly caused by benzophenone. In this scenario, its GWP100 is 17.2 kg CO_2eq per kg octocrylene and, thus, 6.8 times higher than in the reference scenario. This is because in its synthesis, 7.44 kg of benzene are used as input for 1 kg of benzophenone, of which only 0.35 kg react in the synthesis process. Consequently, in this scenario, 7.09 kg of benzene added to the process must be disposed of as hazardous waste. In the scenario assuming 50% reuse, the GWP100 of octocrylene is 14.1 kg CO_2eq per kg, which, still, is twice as high as in the reference scenario (see Figure 6).

These scenarios show that the assumptions on the reuse of the unreacted reactants have a considerable influence on the calculated environmental impacts. On the one hand, it seems unlikely that there is no reuse at all, because it would be a wasteful use of resources. If the rate is as described in the scenarios, the input would (or at least should) be lower, which would also lead to a reduction in the amount to be disposed of, reducing the influence

of the assumption. On the other hand, the process should be conservative in its results. If possible, industrial data should be used that contain exact information about the process.

Third, the assumption on the required thermal energy is analyzed. The sensitivity analysis examines how the results change assuming 1 MJ or 4 MJ instead of 1.8 MJ thermal energy. When 1 MJ is assumed, the GWP100 of octocrylene is 0.07 kg CO_2eq per kg lower than in the reference scenario (−1.0%); if 4 MJ are assumed, it is 0.18 kg CO_2eq per kg higher (see Figure 6). Even when the thermal energy is doubled, the GWP100 of octocrylene only increases by 2.5%. However, the assumption on energy has a higher impact for s-proxies due to the safety factor. If 4 MJ is assumed to be the required energy, the GWP100 of toluenesulfonic acid increases by 31.3% as compared with the reference scenario. However, since only a small amount of toluenesulfonic acid is required for the synthesis of octocrylene, it has little effect on the overall result.

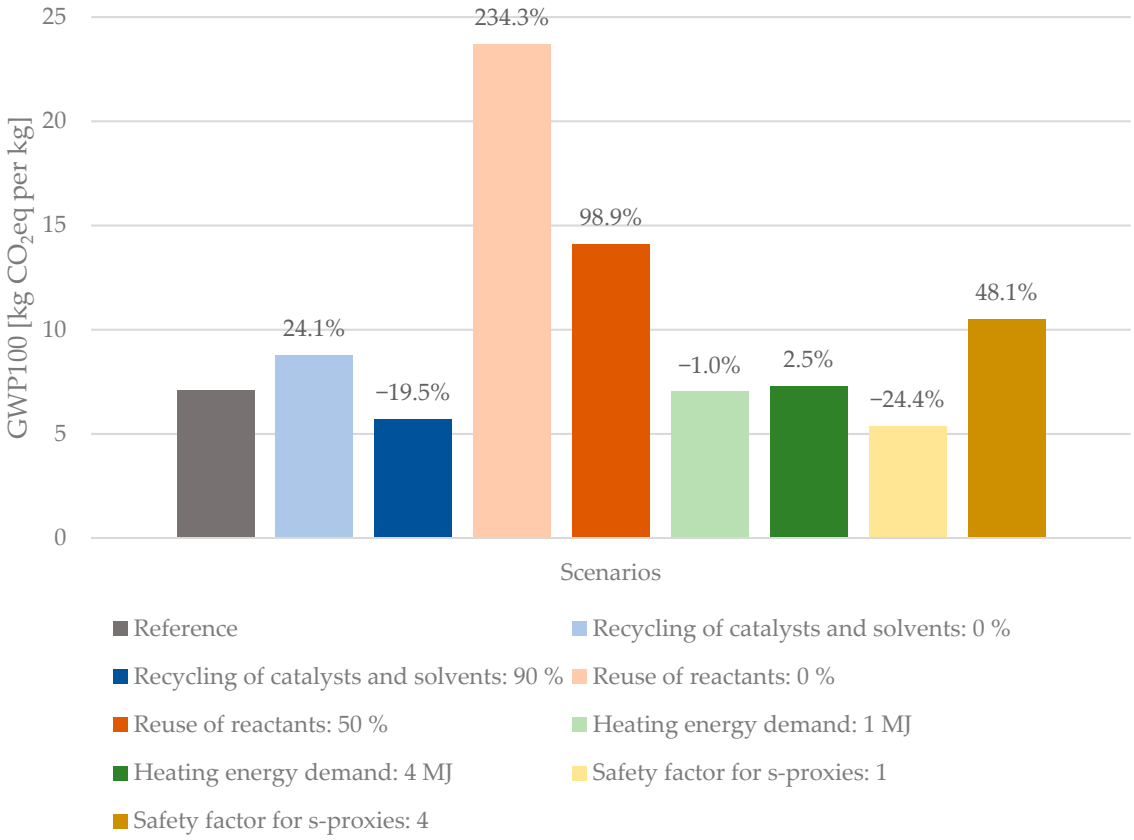

Figure 6. GWP100 of octocrylene in the scenarios considered in the sensitivity analysis as compared with the reference scenario.

The last part of the sensitivity analysis examines the safety factor for s-proxies (ammonium acetate, toluenesulfonic acid, and cyclohexanol as reactant for the synthesis of 2-ethylhexyl cyanoacetate). If a safety factor of 1 is assumed instead of 2, the GWP100 of octocrylene decreases by 24.4% to 5.36 kg CO_2eq per kg (see Figure 6). With a safety factor of 4, the GWP100 of octocrylene increases by 48.1% to 10.5 kg CO_2eq per kg (see Figure 6).

The change is almost entirely due to cyclohexanol, which already causes 69.2% of 2-ethylhexyl cyanoacetate's GWP100 in the reference scenario. Doubling the safety factor and thus the quantity, therefore, leads to an increase of 86.1% in the GWP100 of 2-ethylhexyl cyanoacetate. The influence of toluene sulfonic acid and ammonium acetate on octocrylene's GWP100 is negligible. This means that the influence of the safety factor depends on the overall influence of the s-proxy.

It can be said that the change in the assumptions has a large effect on some substances such as benzophenone, but not on all chemicals modeled for octocrylene. Thus, the influence is strongly dependent on how the process is designed. Therefore, the results, illustrated in Figure 6, are not generally transferable from octocrylene to other chemicals.

Overall, it can be stated that the influence of the thermal energy is less relevant, the safety factor and the recycling of catalysts and solvents have an important influence, but the reuse of reactants affects the result the most.

Validation of the assumptions and of RREM in general should be done by comparing modeled chemicals with ones, where primary data or datasets from LCI databases are available. This is, for example, the case for benzophenone, where a dataset is available in Ecoinvent (octabenzone, synonym for benzophenone) [47]. When comparing the GWP results available in Ecoinvent with 5.06 kg CO_2eq to the one modeled with RREM with 4.66 kg CO_2eq, a difference in the result for GWP of about 8% can be seen. The difference might be explained by applying different chemical reactions and, thus, using different input substances. The Ecoinvent dataset is modeled based on the reaction of 1-bromo-octane and 2,4-dihydroxy-2 benzophenone. In contrast, benzophenone was modeled in this paper with RREM as a reaction of benzene and benzoyl chloride (see Section 3.2). However, a difference of less than 10% shows how well RREM can be used to estimate environmental impacts.

In LCA case studies containing chemicals modeled with RREM, we recommend carrying out an additional sensitivity analysis. Their influence on the overall result of the product system should be investigated by increasing and decreasing their environmental profile by 20%. Hence, the influence of the RREM chemical can be better identified.

6. Conclusions

The RREM-approach introduced in this paper is derived for modeling chemicals in an LCA and is based on the stoichiometry of the synthesis reactions. It enhances the ability to derive datasets that are not yet contained in LCI databases and, therefore, allows for an improved assessment of environmental impacts of chemicals in LCA case studies.

Materials collected during the development of RREM and the application to 60 chemicals are provided (see Supplementary Materials), including Excel® files with summarized data for modeling. The aggregated GaBi datasets of the chemicals listed in Table 1 can be provided upon reasonable request.

Literature data on chemical synthesis usually do not include information on how the production of the considered chemical affects the environment. By linking the inputs and outputs to existing datasets, environmental impacts can be attributed to the chemical and its environmental burdens can be estimated. This offers the possibility of estimating the previously unknown environmental burdens of products that contain chemicals as ingredients, such as cosmetics.

As a follow up, RREM could be further optimized by developing assumptions for additional process components and by validation with available primary data.

Further research could address biogenic substances. Due to the plant origin and, therefore, additional aspects, such as agricultural practices, fertilizer use, etc., RREM cannot be applied one to one, but can be used as a basis to derive overarching rules to model biogenic substances.

Supplementary Materials: The following supporting information can be downloaded at https://www.mdpi.com/article/10.3390/su14063161/s1, Excel® file with an overview of the heat capacities and densities required for the calculations and Excel® file for 60 chemicals (listed in Table 1) with the summarized data needed for modeling datasets.

Author Contributions: Conceptualization, E.H., V.B. and M.F.; data curation, E.H., P.H. and D.B.; formal analysis, E.H.; funding acquisition: V.B. and M.F.; investigation, E.H.; methodology, E.H.; project administration, V.B. and M.F.; supervision, V.B. and M.F.; validation, E.H.; visualization, E.H.; writing—original draft: E.H.; writing—review and editing, E.H., V.B., P.H., D.B. and M.F. All authors have read and agreed to the published version of the manuscript.

Funding: We acknowledge support by the Open Access Publication Fund of Technische Universität Berlin. The study was partially supported by dm-drogerie markt GmbH + Co. KG.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data are contained within the article and the Supplementary Materials.

Acknowledgments: We would like to thank Lena Jarosch and Julia Pelzeter for their support and contributions to data collection.

Conflicts of Interest: The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

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