

Article

Rural Urban Nutrient Partnership (RUN): Life Cycle Assessment of Multi Nutrient Recovery from Kitchen Waste and Blackwater

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Abstract: Newly developed and innovative RUN technology aims to recover nutrients from urban wastewater (blackwater) and biowaste (kitchen waste). The development of RUN technology has been supported by the life cycle assessment (LCA) in order to identify hotspots and trade-offs. While the performance of the process at a laboratory scale did not show any environmental benefits from P recovery, the LCA results have helped to improve the environmental performance at the following scale-up step. The recovery of P on a technical scale was environmentally beneficial, especially in terms of the global warming potential (GWP). However, there were still some trade-offs, e.g., freshwater and marine eutrophication were slightly higher compared to conventional P fertilizer production. Given that P is considered a critical raw material and that climate change is probably the most pressing environmental issue, RUN technology has the potential to deliver on both domains.

Keywords: P recovery; struvite; global warming potential; wastewater treatment; resource scarcity



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1. Introduction

Since the Industrial Revolution, a new era has arisen, the Anthropocene, in which human actions have become the main driver of global environmental change with increasingly negative consequences [1,2]. Human activities have contributed to the exceeding of several of the proposed ‘planetary boundaries’ that define a safe operating space for humankind on a stable Earth system [3–5]. Climate change has already placed approx. 9% of people (>600 million) outside the ‘human climate niche’—defined as people living in regions with an average annual temperature >29 °C [6]. The release of nutrients, particularly nitrogen (N) and phosphorous (P), can cause eutrophication and negatively impacts the quality of freshwater systems. Therefore, wastewater treatment aims to reduce phosphorus and nitrogen emissions to mitigate the risk of eutrophication [7].

The use of fertilizer is essential for sufficient food and feed production. The demand for nitrogen (N), phosphorus (P) and potassium (K) can just partially covered from manure and crop residues and must be complemented by energy-intensive N-fertilizer and resource-constrained mineral fertilizers such as potassium salts and rock phosphate [8,9]. However, fertilizer production and application cause various environmental impacts [10] therefore their production processes should be further optimized in environmental terms. This is even more relevant for alternative nutrient recovery processes.

Life Cycle Assessment (LCA) is a suitable tool to aid relevant technology development for those technologies at an early stage [11]. Investigating nutrient recovery from wastewater or sewage sludge using LCA is common practice affirmed by several literature studies and reviews [12–15].

As part of the Green Deal Klicken oder tippen Sie hier, um Text einzugeben. (https://commission.europa.eu/strategy-and-policy/priorities-2019-2024/european-green-deal_en,

accessed on 23 February 2024), the Farm to Fork strategy (https://food.ec.europa.eu/horizontal-topics/farm-fork-strategy_en, accessed on 10 February 2024) further emphasizes sustainable food production in the context of a circular bio-based economy using fewer mineral fertilizers and plant protection agents. Phosphorus cannot be produced synthetically or replaced [16]. The European Union (EU) is almost entirely dependent on phosphate imports. Due to this dependence on imports, the EU classified P as a 'critical raw material' (https://single-market-economy.ec.europa.eu/sectors/raw-materials/areas-specific-interest/critical-raw-materials_en, accessed on the 23 December 2023), and wastewater has emerged as a viable resource for P recovery [17–19].

The German Sewage Sludge Ordinance (AbfKlärV) forces a paradigm shift upon the operators of wastewater treatment plants (WWTPs). For WWTPs with a population equivalent (PE) of more than 50,000, the application of untreated sewage sludge to soil is prohibited. The threshold value above which the AbfKlärV stipulates a legal obligation to recover phosphorus from sewage sludge is 20 g P/kg DM. The latter applies regardless of the size of the sewage treatment plant. The co-incineration of sewage sludge in cement plants, waste incineration plants, etc., will only be permitted for sewage sludge with less than 20 g P/kg DM. This means that the construction of expensive, resource-intensive sewage sludge mono-incineration plants as well as increased sludge transportation will be necessary.

Heavy metals are concentrated in the sludge and their concentration depends on the origin of wastewater [20]. In general, the contamination level of recycled nutrients must be kept to a minimum [21]. Nutrient recovery also has an impact of the environment due to the use of energy and chemicals, which can lead to a problem shift rather than an overall benefit [22]. The collection of blackwater (urine + feces + flushing water + toilet paper) together with kitchen and garden waste at their source could allow for the recovery of nutrients [23,24] without cross-contamination from other wastewater streams, e.g., industrial wastewater.

The collection and treatment of household wastewater and biowaste using various technologies in combination enable not just the recovery of nutrients but also the utilization of organic carbon as the substrate for biodegradable biopolymer (polyhydroxyalkanoates (PHA)), biochar, or biogas production.

The cascade treatment of household wastewater and biowaste is in line with the European Bioeconomy Strategy and the Circular Economy Action Plan that advocate the recovery of nutrients from waste streams and should be part of an Integrated Nutrient Management Plan [25]. Recovered nutrients from urban wastewater can be reused in neighboring rural regions as a starting point of a rural urban nutrient partnership (RUN).

RUN technology (<https://en.run-projekt.de>) was developed on several scales as part of the RUN research project. Using RUN technology, the nutrient-containing material streams of kitchen waste and blackwater were combined in defined mixing ratios and treated mechanically, physico-chemically, and biologically. Phosphorus, nitrogen, and potassium contained in the waste streams were mobilized and refined. The aim was to recover the nutrients as effectively and sustainably as possible and to refine them into marketable products. For example, desired volatile fatty acids were produced in bioreactors and the compositions of nutrients were controlled in physico-chemical processes. Innovative separation techniques contributed to the efficient separation of solid and liquid fractions or the retention of pollutants. Liquid fractions were used to produce complex fertilizers and organic carbon was used as feedstock for bio-based plastics [26]. Solid fractions were used to produce biogas or biochar as a nutrient carrier and soil conditioner. The efficient collection and separation of waste and wastewater streams at the 'households' meant that, in contrast to conventional wastewater treatment (central municipal sewage treatment plant), the recycling products were not contaminated with wastewater-borne commercial and industrial pollutants. Decentralization, as a key feature of the RUN concept, generates environmentally relevant advantages. The organic waste and wastewater were directly transported to the RUN pilot plant via vacuum systems, saving valuable drinking water

as a transport medium. The great potential of RUN technology lies both in the efficient treatment of relatively small quantities with high nutrient concentrations and in the avoidance of long transportation routes for both the wastewater and waste streams generated in residential areas and the recycling products for use in agriculture near residential areas.

The potential environmental implications of the applied technologies (for struvite ($MgNH_4PO_4$) precipitation, ammonium, and potassium recovery, and PHA-enriched biomass) at laboratory and pre-pilot scale (hereafter referred to as ‘technical scale’) were assessed using life cycle assessment in order to optimize the technical processes as well as identify trade-offs and environmental hotspots along the treatment chain. The applied technologies were assessed from a P recovery perspective. Previous studies [27–30] consider waste and wastewater treatment as service and use an input-based functional unit; however, in this case, the recovery of nutrients and other value-added products is the dominant aim. RUN technology delivers multiple products; consequently, a product perspective is used to assess the technology development.

The objective of this paper is to estimate the environmental implications of P recovery from a mixture of kitchen waste and blackwater on a life cycle basis using RUN technology.

2. Results

2.1. System

The treatment of kitchen waste (KW) together with blackwater (BW) with RUN technology at laboratory scale (1 L) and technical scale (110 L) was assessed in this paper. The composition of blackwater and kitchen waste is provided in the Appendix A (Table S1).

RUN Technology encompasses a cascade of various processes with the aim to treat the combined biowaste and blackwater in order to reduce environmental burdens and recover nutrients and other value-added substances simultaneously before they are discharged to the WWTPs and diluted there. The initial process scheme of RUN technology is shown in Figure 1, and the modified process scheme after optimization is shown in Figure 2.

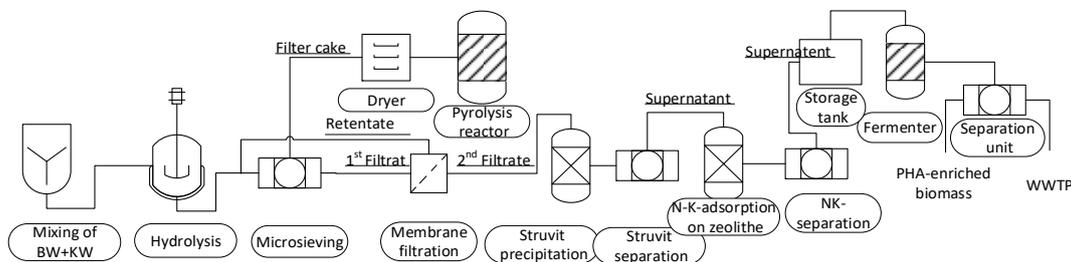


Figure 1. Process scheme of RUN technology at laboratory scale.

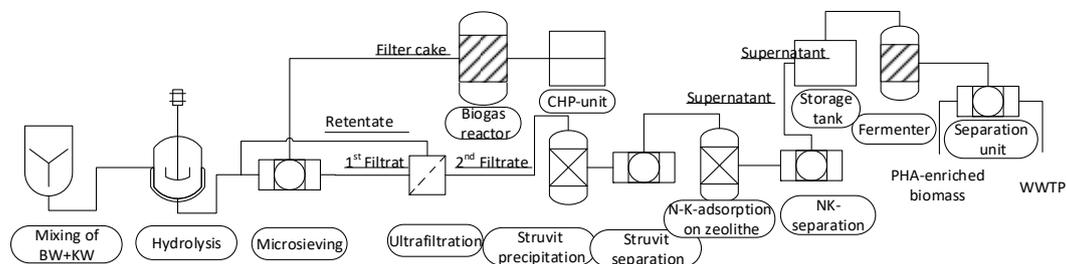


Figure 2. Process scheme of modified RUN technology at technical scale.

After hydrolysis and the first filtration process (microsieving, pore size $0.03\ \mu m$), the solid fraction (filter cake) was dried and processed to biochar, while the filtrate was treated with a second filtration process (ultrafiltration). The retentate was returned to hydrolysis. The permeate was used to precipitate struvite with magnesium chloride hexahydrate. After separating other value-added products, the remaining wastewater was treated together with the household’s grey water in conventional WWTPs (not shown in the process scheme).

For the conventional wastewater treatment and the calculation of avoided burdens, Ecoinvent datasets were used.

Kitchen waste was shredded and mixed with blackwater. At laboratory scale, sulfuric acid (7.5–12.2 mmol per L permeate) was added to transform organic-bound phosphorus to dissolved hydrogen phosphates. After filtration, followed by ultrafiltration, the filter cake was used for producing biochar. Magnesium chloride hexahydrate (for the LCA, MgO equivalents were used instead of $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ because the latter is not available in Ecoinvent and both share the initial process steps; the same assumption is applied in the following analysis (<https://www.umweltbundesamt.de/publikationen/oekobilanzieller-vergleich-der-p-rueckgewinnung-aus>, accessed on the 15 July 2023)) (0.64–0.76 g per L permeate) and sodium hydroxide (15.3–49.0 mmol per L permeate) were added to the permeate in order to precipitate struvite. After struvite separation, zeolite (120–240 g per L permeate) was added to the filtrate in order to recover ammonium and potassium. The remaining filtrate was used for PHA production; thus, the remaining carbon was also utilized to produce a value-added product. The processing of the PHA-enriched biomass to purified PHA was not conducted within this project; instead, data from the literature were used to estimate the associated environmental impacts.

At technical scale, an anaerobic bioreactor was used instead of sulfuric acid in order to hydrolyze the mixture. During hydrolysis, nutrients were released, and volatile fatty acids (VTAs) were produced. Compared to the laboratory scale, the consumption of operating materials was substantially reduced: 0 mmol per L sulfuric acid permeate; 0.58 g magnesium chloride hexahydrate (solid) per L permeate; 14.4 mmol sodium hydroxide per L permeate; and 131 g zeolite (solid) per L permeate. The filter cake produced during the subsequent solid–liquid separation was used as biogas substrate instead of being processed to biochar.

With the modified RUN technology, it was possible to recover 60% P, 41% K, and 30% N from the blackwater and kitchen waste mixture, thereof approx. 56% of P as struvite and 4% adsorbed on zeolite together with N and K. Moreover, approx. 35% P remained in the filtercake, which was used for biogas production. The mass flow diagram of P along the process chain is provided in the Appendix A (Figure S1).

In total, approx. 95% P can be recovered by RUN, assuming that the digestate of the biogas plant is suitable to be used as an organic fertilizer.

2.2. Identification of Substitution Products

RUN technology generates multiple products. The procedure described by [31] was used to identify substitution products. For the sake of simplicity, magnesium (Mg) was not considered as recovered as it was added to the process in order to precipitate struvite; consequently, no credit was given for Mg. Moreover, the MgO dataset was used instead of MgCl_2 , although the latter has less environmental impacts. Polylactic acid (PLA) was assumed as substitution product for unpurified PHA due to limited data availability. Both assumptions increased the uncertainty of the results, however that approach follows the precautionary principle and thus environmental impacts were not underestimated. The products identified for substitution were mineral fertilizers and PLA. Mineral fertilizers in Germany that would be replaced in the market were identified using the marginal product principle.

Calcium ammonium nitrate and ammonium nitrate were identified as marginal N fertilizers for Denmark by [32,33]. Ammonium nitrate was also identified as a marginal N fertilizer for The Netherlands [34] and Sweden [35,36].

Marginal products were selected by using the approach defined by [33]. The technology with the lowest long-term production cost were considered marginal. Looking at the market trends for mineral fertilizers in Germany, a decreasing demand over the past decade was observed, and the same is forecasted for the forthcoming decade. Hence, the least competitive mineral fertilizer for the respective nutrients is the marginal product. The fertilizer data available in the Ecoinvent database was considered for the marginal fertilizer [31], as shown in Table 1.

Table 1. List of single-nutrient fertilizer datasets available in the Ecoinvent database. The respective nutrient contents are given in brackets (adapted from [31]).

N Fertilizer	K Fertilizer
Ammonium Nitrate (35% N)	Potassium Chloride (60% K ₂ O)
Ammonium Sulphate (21% N)	Potassium Sulphate (50% K ₂ O)
Calcium Ammonium Nitrate (26.5% N)	
Calcium Nitrate (12% N)	
Urea Ammonium Nitrate (32% N)	
Urea (46%)	

2.3. LCA Results at Laboratory and Technical Scale

Results for the product perspective (1 kg P₂O₅ recovered) at laboratory and technical scales are shown in Table 2. The columns categorized as ‘total incl. credits’ show the final results after the substitution of products other than P. The columns categorized as ‘chemicals’ show the contribution of used operating materials with RUN technology.

Table 2. Environmental impacts at laboratory scale and technical scale per kg P₂O₅ recovered.

Impact Category	Reference Unit	Lab Scale Total Incl. Credits	Lab Scale Chemicals	Tech Scale Total Incl. Credits	Tech Scale Chemicals
Fine particulate matter formation	kg PM _{2.5} eq	2.0×10^{-1}	1.5×10^{-1}	7.1×10^{-2}	3.5×10^{-2}
Fossil resource scarcity	kg oil _{eq}	7.3×10	2.8×10	5.2	1.4×10
Freshwater ecotoxicity	kg 1.4-DCB	2.0×10	1.5×10	-1.3×10	2.5
Freshwater eutrophication	kg P _{eq}	1.6×10^{-1}	1.4×10^{-1}	1.3×10^{-2}	2.8×10^{-2}
Global warming (GWP)	kg CO ₂ eq	7.3×10	1.2×10^2	-9.8	5.4×10
Human carcinogenic toxicity	kg 1.4-DCB	1.1×10	9.5	1.5	3.2
Human non-carcinogenic toxicity	kg 1.4-DCB	3.2×10^2	3.1×10^2	-7.4×10	9.7×10
Ionizing radiation	kBq Co-60 eq	9.8	1.6×10	2.2	1.7
Land use	m ² annual crop _{eq}	-1.8×10	4.4	-8.4	-2.5
Marine ecotoxicity	kg 1.4-DCB	2.5×10	1.9×10	-1.6×10	3.5
Marine eutrophication	kg N _{eq}	1.2×10^{-2}	1.5×10^{-2}	1.3×10^{-3}	2.2×10^{-3}
Mineral resource scarcity	kg Cu _{eq}	7.3×10^{-1}	6.1×10^{-1}	-3.6×10^{-1}	1.5×10^{-1}
Ozone formation. Human health	kg NO _x eq	1.7×10^{-1}	1.6×10^{-1}	-1.1×10^{-1}	4.7×10^{-2}
Ozone formation. Terrestrial ecosystems	kg NO _x eq	1.8×10^{-1}	1.7×10^{-1}	-1.1×10^{-1}	4.8×10^{-2}
Stratospheric ozone depletion	kg CFC11 eq	-2.9×10^{-4}	1.5×10^{-4}	-1.1×10^{-3}	3.6×10^{-5}
Terrestrial acidification	kg SO ₂ eq	4.8×10^{-1}	3.3×10^{-1}	2.3×10^{-1}	6.2×10^{-2}
Terrestrial ecotoxicity	kg 1.4-DCB	4.8×10^2	5.3×10^2	-5.2×10^2	1.0×10^2
Water consumption	m ³	-1.3×10	3.0	1.6	1.3

At the laboratory scale, the recovered P as struvite had a GWP of 73 kg CO₂ eq/kg P₂O₅ compared to approx. 1–2 kg CO₂ eq/P₂O₅ for the industrially manufactured fertilizer, depending on the kind of P fertilizer (Ecoinvent datasets for industrially produced P fertilizer). The main contributors to GWP were sodium hydroxide and the heat required for drying the filter cake. This result shows the importance of investigating the environmental impacts of phosphorous recovery processes, which is also in agreement with [37].

The fossil resource scarcity followed the pattern of the GWP. The energies required for drying the filter cake, NaOH, and zeolite were the main contributors to the impact category fossil resource scarcity, in this order. Drying the filter cake dominated fossil resource scarcity, while the credits from other products could just compensate the impacts of NaOH and zeolite. All environmental impacts of P recovered from backwater and kitchen waste at laboratory scale were substantially higher than those of conventional mineral P fertilizers.

The hotspot analysis, shown in Figure 3, revealed that the added chemicals and the drying of filter cake were the most dominant contributors to relevant impact categories at the laboratory scale. Therefore, the hydrolysis process was modified, and the filter cake was used as the substrate for biogas production. Biogas was used in a virtual CHP plant, and the generated heat from the CHP was used within the process chain, while the system was credited for the electricity generated from biogas using a German supply mix. Sulfuric acid was eliminated and this resulted in a reduced demand of NaOH for adjusting the pH.

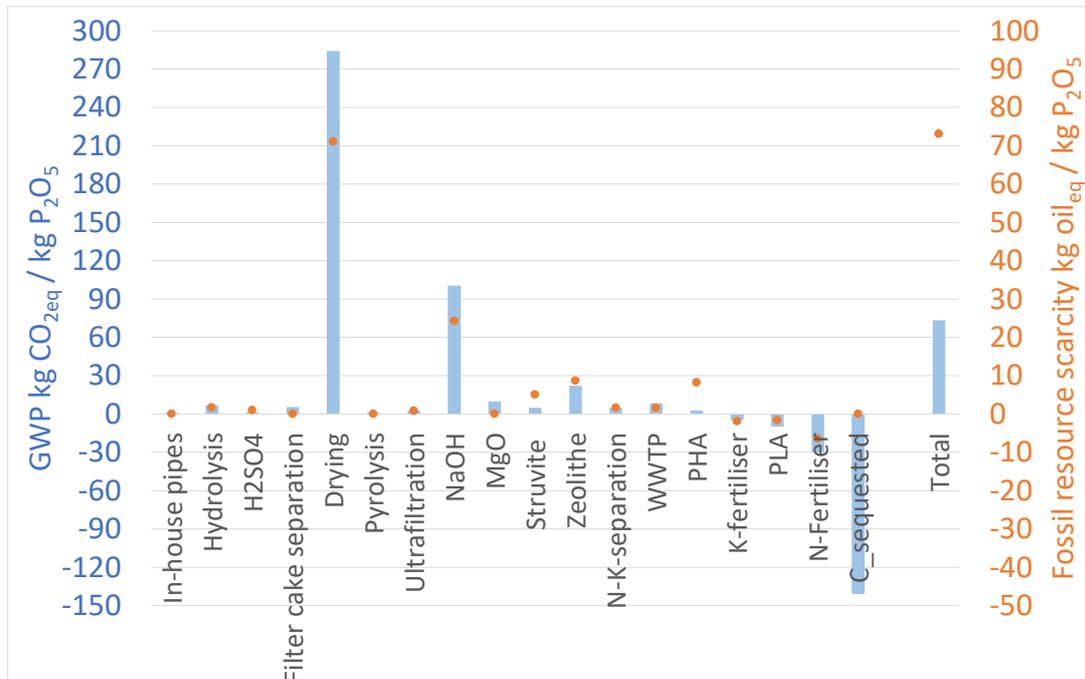


Figure 3. GWP and fossil resource scarcity per kg P₂O₅ recovered at laboratory scale.

These amendments were implemented in the scale-up step. Results of GWP and fossil resource scarcity for the amended process chain at technical scale are shown in Figure 4.

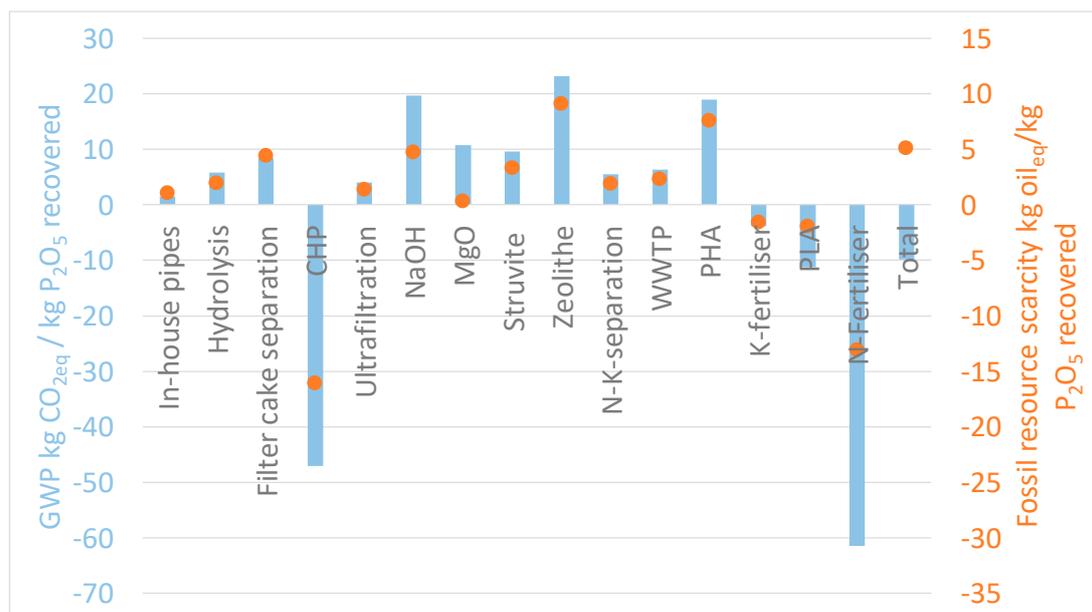


Figure 4. GWP and fossil resource scarcity of recovered P₂O₅ at technical scale.

3. Discussion

The main contributors to the GWP of the P_2O_5 recovered at laboratory scale were NaOH and the heat required for drying the filter cake. The heat gained during the pyrolysis process could not compensate for the heat required for drying. The value shown in Table 2 and Figure 3 is the net heat requirement, i.e., total heat demand minus regained heat from pyrolysis. The main credit with respect to the GWP would come from C sequestration if the biochar was used as a soil improver. For this purpose, it was assumed that 80% of the carbon was sequestered. Biochar can be a large and long-term carbon sink when applied to soils [38]. While the composition and quality of biochar vary, those produced under moderate to high temperatures have stable C that is likely to persist for hundreds of years [39]. According to the IPCC, 80% ($\pm 11\%$) of the C present may be stable and considered permanently sequestered after 100 years [40]. This assumption was used to estimate the credits due to C sequestration. Although this is a simplification, any reasonable change would not change the overall picture. Originally, biochar was also foreseen as an adsorbent for N-K adsorption, but its performance was not convincing at the time.

RUN technology's recovery of P at a laboratory scale did not show notable environmental benefits compared to the conventional production of the P fertilizer, such as single or triple super phosphate. At laboratory scale, the majority of environmental impacts can be attributed to the use of chemicals, particularly sodium hydroxide. Even credits given for N and K recovery as well as for bioplastic (using PLA as a dummy for PHA) and C sequestration due to biochar application do not change the overall picture. All environmental impacts of the recovered P were higher than those of the P fertilizer available on the market, compared to the environmental impact values given in the Ecoinvent database for single and triple super phosphate. The contribution of each process step to the GWP and fossil resource depletion is shown in Figure 3. The patterns of contributors for the other environmental impact categories are quite similar and are therefore not discussed in detail.

At technical scale, the GWP of the recovered P_2O_5 was negative, mainly due to the credits received from the electricity produced by CHP and the production of the N fertilizer. Both are also responsible for most of the credits related to fossil resource scarcity, but, for this impact category, the credits could not compensate the fossil resource demand for producing zeolite, PHA-enriched biomass, and NaOH.

With respect to other environmental impacts, recovering the P fertilizer at technical scale showed a mixed picture. Human carcinogenic toxicity, ionizing radiation, eutrophication, and fossil resource scarcity in particular were higher than those from conventional P fertilizers (results for conventionally produced P fertilizer are taken from the Ecoinvent database; the results are available for all with a Ecoinvent license). Although the recovery of the P fertilizer from kitchen waste and blackwater has numerous environmental benefits, there were still some trade-offs at the current technology development stage. Most of them were not related to a direct contribution of the process chain but due to the production of chemicals used as operating materials in RUN technology, particularly NaOH as well as zeolite, and, in the case of human carcinogenic toxicity, due to hexavalent chromium exposure during MgO production. An exception was the production of PHA. PHA production with RUN technology was energy-intensive, and the required heat from natural gas was a substantial contributor to fossil resource depletion. RUN technology is not fully developed and there might be further environmental benefits to come. Improvements of the environmental performance of RUN technology can be achieved by reducing the demand of NaOH or by replacing zeolites for N-K adsorption from the filtrate with more environmentally friendly adsorbent materials.

There are various LCA studies on nutrient recovery from wastewater [12,22–24], with all of them using different feedstock compositions, process parameters, and methodological choices. Therefore, comparing the results of this study to other literature studies is not possible without harmonizing functional units, system boundaries, feedstock compositions, and a number of methodological choices, among others. This is commonly recognized in

the LCA community. A reasonable comparison of LCA results will only be possible if the procedure described in ISO 14044 [41] for comparative assertion is applied.

4. Materials and Methods

Treating biowaste together with blackwater was used to recover nutrients and other value-added products. The main drivers for developing a new technology were P scarcity and the legal requirements to recover P from sewage sludge or sewage sludge ash. One promising solution is to separate P-rich streams as far as possible before they reach a municipal wastewater treatment plant [30]. The collection takes place directly at households, and the collected blackwater and biowaste are processed using the RUN treatment chain.

LCA provides a holistic view that quantifies the environmental impacts of a product, process, or system by tracking the material and energy flows from raw material extraction through manufacturing and use to the end of life [41,42].

This study assesses a process combination that is not yet implemented in full scale, although some process steps are. The LCA was conducted based on ISO 14040 and 14044. The systems were modelled in openLCA 2.04 (<https://www.openlca.org/>), and the Ecoinvent database 3.9.1 (<https://www.ecoinvent.org>) was used for background processes.

Background processes were chosen to be representative for Germany or, if not available, the following order was applied datasets for Europe and then datasets globally average datasets. The data for foreground processes were directly measured from partners of the RUN project. The RUN technology under investigation is still at the development stage but may be applied at large scale in the coming decade. Therefore, a consequential approach was used.

ReCiPe 2016 [43] was used as the impact assessment method, and the hierarchist (H) perspective was selected. All impact categories were calculated, but just the most relevant ones are discussed in detail. Data of foreground processes was measured/collected in the RUN research project; hence, the selected ReCiPe perspective was considered most appropriate; this is also in agreement with [44].

System Boundary

The system encompassed the collection, treatment, and recovery of nutrients and other products; hence, it represented a cradle-to-gate system. The possible processing into multifunctional fertilizers and the application of the recovered nutrients were outside the system's boundary.

The system was modelled based on the total annual amount of waste water and kitchen waste of one inhabitant in the city of Heidelberg in Germany. The combined amount to be treated with RUN technology was approx. 3.7 m³ in addition to 39 m³ grey water per person per year in a conventional WWTP. Given that P recovery was the main driver for RUN technology development, one kg of the recovered P fertilizer as P₂O₅ was used as functional unit.

5. Conclusions

LCA proved to be a very powerful tool for identifying and improving the environmental performance of RUN technology. Critical factors influencing the global warming potential could be identified and used as starting points for optimizing technical details. Thus, the first upscaling step from the laboratory scale to the technical scale resulted in substantially lower environmental impacts. The ongoing upscaling process of RUN technology to pilot scale could therefore lead to an even better environmental performance, especially if the consumption of energy and other operating resources related to N, P, K, and C recovery is improved.

At technical scale, the recovery of P from kitchen waste and blackwater led to a substantial reduction of greenhouse gases compared to the production of mineral P fertilizers. However, there were also trade-offs, e.g., freshwater and marine eutrophication of RUN technology were slightly higher compared to conventional mineral P fertilizers. Although

this is a drawback, it can be accepted because the marine and freshwater eutrophication due to fertilizer application are substantially higher than those caused by its production. The efficient use of P fertilizers is more relevant for freshwater eutrophication than for the P production process.

Given that P is considered a critical raw material and that climate change is probably the most pressing environmental issue, RUN technology has the potential to deliver on these two domains.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/recycling9020031/s1>, Figure S1: Mass flow diagram of P in RUN, Table S1: Composition of blackwater and kitchen waste.

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Data Availability Statement: The authors confirm that the data supporting the findings of this study were measured within the RUN project. The raw data of this study are available from the authors upon reasonable request.

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

Appendix A

The appendix contains data on the composition of blackwater and kitchen waste used for the experiments as well as a mass flow diagram of P along the process chain (Table S1).

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