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Environmental Prospects for Mixed Textile Recycling in Sweden

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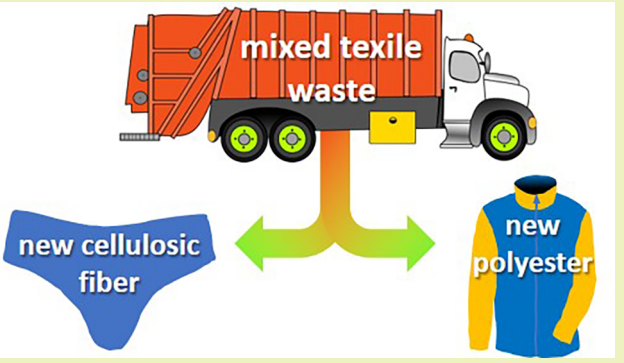
Supporting Information

ABSTRACT: The production of cotton and other fibers causes excessive resource use and environmental impacts, and the deployment of these fibers in "fast fashion" is creating large masses of textile waste. Therefore, various industrial researchers are attempting to develop systems to recycle cellulosic materials. This is a challenging undertaking because of the need to handle mixed waste streams. Alkaline hydrolysis has been suggested as a useful textile recycling process, but its sustainability credentials have not been fully examined via life cycle assessment. The aim of this article is to provide such an examination and to guide process developers by scaling up results from recent laboratory work to a small-scale industrial facility. The results indicate that the recycling process is promising from an environmental point of view. The key issue controlling the relative environmental performance of the recycling system in comparison to a single-use benchmark is how the process for converting recovered cotton into a cellulosic fiber is performed. A fully integrated viscose production system or a system that makes one of the newer cellulosic fibers (e.g., lyocell) from the recovered cotton will improve the performance of the recycling system relative to its alternatives.



KEYWORDS: *Life cycle assessment, Polyester, Cotton, Chemical recycling, Fiber recycling, Alkaline hydrolysis*

INTRODUCTION

Our rising global population, increased affluence, and the phenomenon of "fast fashion", in which the garment industry drives rapid processes from design to delivery and produces less durable clothing, have led to a dramatic increase in the production of textiles. As shown in Figure 1, global per capita textile fiber production has increased from 5.9 kg in 1975 to 13 kg in 2018.[1] This growth in textile production is associated with environmental challenges including the demands placed on natural resources, emissions associated with textile production and use, and the generation of solid waste. In the United States, approximately 40 kg of textile waste is generated annually per person; for China, the corresponding figure is 19 kg.[2] Sweden is a wealthy country in which the consumption of textiles has reached 22 kg per person and year.[3] In Sweden and most countries, the majority of used textile products are destined for either landfill or incineration with energy recovery.[2]

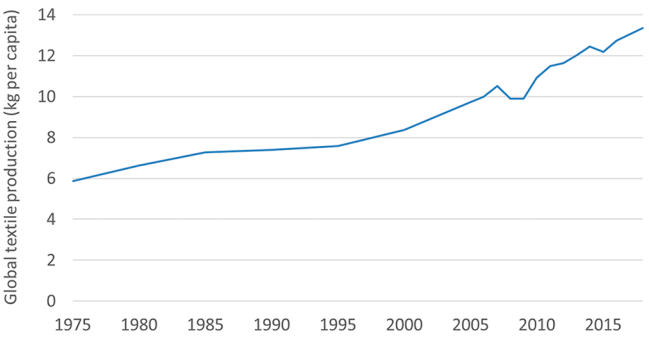


Figure 1. Global per capita textile fiber production 1975−2018.

While energy recovery from textile waste is beneficial, recent studies quantitatively indicate that greater benefits may be available via reuse of garments, reuse of textile materials, or the dissolution of their polymeric structures for up-cycling the waste.[4−6]

Textile recycling has the potential to reduce the use of natural resources and the generation of textile waste, but there are significant challenges to the development of textile recycling systems. One of the key challenges is that textiles are damaged by use and laundering, not just at the level of the fibers but also at the level of their constituent polymer chains,[7] which means that mechanical material recycling is at best a down-cycling process with products exhibiting reduced fiber strength. So while mechanical recycling systems are the predominant form of textile recycling today, they have limited potential to supply alternatives to the raw materials in use by the garment industry.

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Another key challenge is the need to provide a reasonably pure stream of material for recycling, given the wide range of physical contaminants (metallic zips, plastic buttons, etc.) which may be present in collected textile waste, and the blend of different polymers used to make garments (e.g., elastane). The latter challenge has confined commercial up-cycling systems to relatively pure recycle streams. One example is the development of open and closed loop recycling systems for polyester-rich recyclates by the company Teijin, which claims to be constructing the world's largest polyester recycling facility in China to process 25,000 tonnes of waste polyester per annum.[8] The intention is to use textile and packaging waste (e.g., PET bottles) to create a reliably polyester-rich inlet flow. Another example is the opening in 2017 of the Re:newcell pilot facility in Kristinehamn, Sweden, with a capacity of 7000 tonnes per year.[9] This facility is intended to accept a cotton-rich recycle stream.

The availability of relatively pure recyclable material streams is a bottleneck to the use of facilities like these to recycle textile waste,[10] so there is therefore a need for recycling systems that can up-cycle mixed wastes. Most of the processes described in a recent review of textile recycling chemistries concerned the production of products other than textile fibers,[2] for example ethanol can be produced by fermentation of solutions derived from acidic or enzymatic degradation of cotton in a mixed textile stream. The use of ionic liquid solvents is also being examined, although environmental hazard and cost considerations have hindered their uptake.[2] In order to facilitate scale-up and adoption of mixed textile recycling by existing cellulose-industrial facilities, Palme et al.[11] and Björquist[12] focused on alkaline degradation of polyester in a mixed textile stream into its component parts (terephthalic acid and ethylene glycol). The key operations and principal material flows in the overall recycling process [called "Blend Re:wind" (BRW) by de la Motte and Barnekow][13] are illustrated in Figure 2. In principle, this process has the advantage that it uses common and inexpensive chemicals and also that it can provide a cellulosic material suitable for valorization via existing forest cellulosic fiber production facilities. However, like most of the processes available today at the laboratory scale, the environmental performance of the process has not been subject to a peer-reviewed life cycle assessment (LCA), which is necessary for policy-makers and investors to understand whether the demand for energy and chemicals caused by recycling processes outweighs the benefits of recycling[2] and therefore whether it is worthwhile to support the development of the method and its implementation in Sweden and around the world. This paper aims to examine the potential environmental sustainability of the BRW process in comparison to current benchmarks and to provide input to the design of future recycling infrastructure for blended fiber materials.

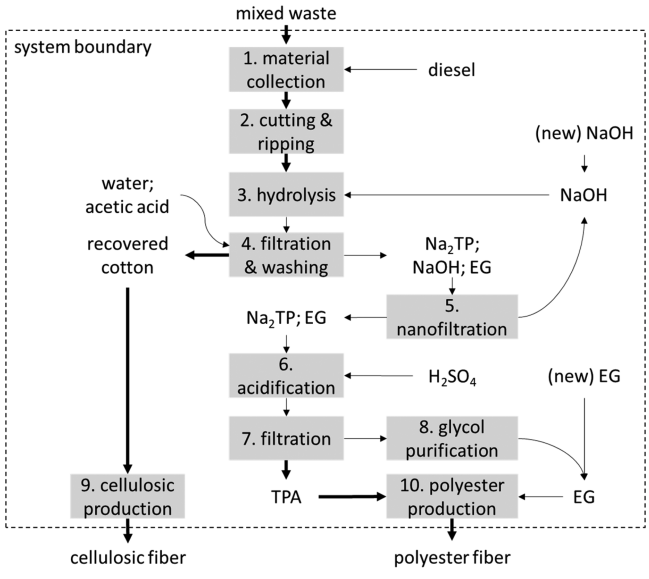

Figure 2. System diagram for BRW scenario (thicker arrows indicate solid phase flows. Na_2TP = sodium terephthalate; EG = ethylene glycol; and TPA = terephthalic acid).

METHOD

Scenario Construction. The functional unit (analytical basis) of this LCA is the recycling of 850 tonnes of mixed textile waste. This value is our forward estimate of the amount of mixed textile waste available in 2023 from commercial laundries that manage textiles for the Swedish healthcare sector (based on Brismar, 2014). While this represents a small scale facility, it is considered to offer a relatively consistent flow of materials and an appropriate scale of operations for industrial developers needing to test upscaling the BRW process from current laboratory data prior to proceeding to a full-scale industrial facility.[14]

An overview of the BRW recycling scenario is provided in Figure 2. Details regarding the unit operations numbered 1−10 are provided in the Supporting Information. Briefly, the figure shows the collection of mixed textile waste to a national facility where the waste is shredded and mixed with a sodium hydroxide solution. A solid flow of recovered cotton is filtered from the solution of sodium terephthalate (Na_2TP) and ethylene glycol (EG). The cotton is washed in acid and then neutral water. The solution is acidified to precipitate solid terephthalic acid (TPA), resulting in a salty liquid which can be distilled to separate the ethylene glycol product. The terephthalic acid and ethylene glycol can be subsequently combined to make new polyester. Energy and mass balances for these processes were developed by scaling up results presented by Palme et al.[11] and Björquist[12] using fundamental chemical engineering data, scientific literature, dialogue with industry, and ultimately numerical simulation in a Microsoft Excel environment. Data on the production of the energy and chemicals was primarily obtained from the Gabi Professional database (version 8.7, service pack 36), developed by Thinkstep. In cases where additional LCI information was required on background processes, this was complimented with some data from the current Ecoinvent database (version 3.5) also provided by Thinkstep. The Gabi software was used for modeling the product system and calculating the life cycle impact assessment (LCIA) results.

A recycling scenario in which ethylene glycol is not recycled was also included in the analysis because the purification process appeared to be an environmental hotspot in initial calculations when the starting solution is as dilute as Björquist[12] suggests (about 0.6% ethylene glycol from a 3% waste suspension; note that we drive the predistillation glycol concentration above 2% in our model using the nanofiltration step). In this scenario, virgin ethylene glycol is produced for combination with the recycled terephthalic acid, while the waste stream containing unrecycled ethylene glycol is eliminated via a wastewater treatment facility. This scenario is illustrated in Figure 3. These are cradle-to-gate scenarios in the sense that the system boundary reaches from raw material in the environment (or low-value waste textile) to the production of textile fiber ready for spinning into yarn. Consistent with many other studies of textile recycling (see Sandin and Peters),[6] cutoff allocation has been applied to the recyclable material used in the BRW scenarios, and it is assumed that the recycled fibers are of sufficient quality to replace fibers produced from virgin resources in a 1:1 ratio.

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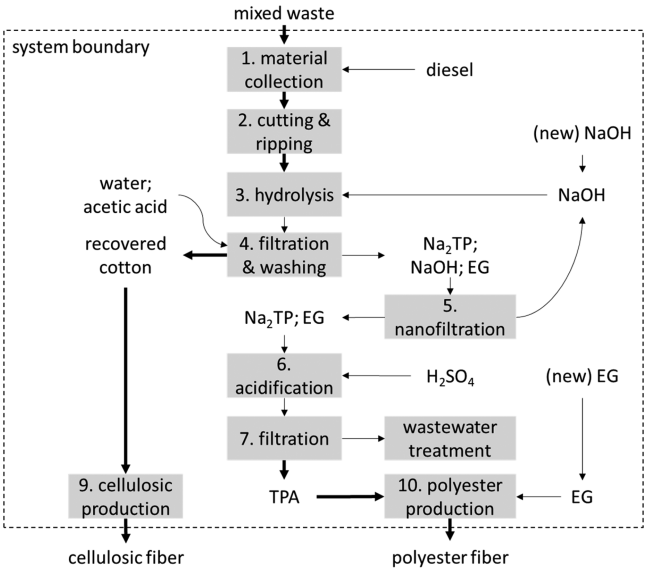

Figure 3. System diagram for BRW scenario without recycling of ethylene glycol ("BRW ex EG").

Since there is limited potential to increase global cotton production, the principal alternative to the BRW process for the production of cellulosic fibers is the use of forest resources. The dominant forest cellulosic fiber today is viscose, therefore the BRW process is modeled as producing viscose and recycled polyester. For a business-as-usual benchmark, we developed a single-use scenario in which mixed textile waste is incinerated (making it an energy resource rather than a material resource) in parallel with the production of virgin polyester (vPES) and viscose (from forest resources). A fourth scenario was included to represent the single-use of cotton instead of viscose, because cotton is currently the dominant cellulosic fiber in the global market. These two single-use scenarios are illustrated in Figure 4. These scenarios were modeled using cradle-to-gate data from the Gabi Professional database and are described in more detail in the Supporting Information.

Sensitivity and Uncertainty Analysis. Initial model iterations indicated the relatively high significance of the production process for viscose from wood pulp and the production of cotton fiber. The cotton fiber data is relevant to one of the single-use scenarios. The viscose production data is used in both a single-use scenario and in the BRW scenarios (box 9 in Figure 2). We were also interested in the potential for the selection of cellulosic fibers other than viscose to reduce the environmental impact of both the benchmark single-use scenario and the BRW scenarios (for example lyocell and modal) but decided that the performance of the alternatives is within the range of environmental performance of different viscose facilities (see the Supporting Information). Therefore, we adopted the approach to uncertainty analysis of varying the emissions and resource demands of the cellulosic fiber production processes by a factor of 0.5 to 2.0 to reflect the uncertainty in both the underlying data for the particular cellulosic fiber production process and the uncertainty in the scope regarding the potential choice of cellulosic fiber.

Life Cycle Impact Assessment. LCIA methods were chosen in order to reflect the principal environmental impacts of the textile industry, as identified in previous reports.[15] The impacts considered are acidification potential, climate change, eutrophication, freshwater ecotoxicity, human toxicity (cancer and noncancer), primary energy use, and water scarcity. The characterization models used to calculate the scores for these impacts are drawn from the European Commission's Product Environmental Footprint program released in May 2018[16] as implemented in the Gabi software[17] with the exception of the primary energy indicator which is based on the consumption of both fossil and nonfossil resources (as implemented in Gabi). In this way, the energy use indicator reflects a concern not just for the depletion of fossil resources (which for our scenarios is in any case strongly reflected in the climate change indicator) but also the equitable sharing of energy resources among contemporary needs. We considered including a land-use indicator (i.e., LANCA),[18] but while the original method has been substantially revised in response to perceived shortcomings, the current version has not been implemented in the Gabi software, so we omitted it.

RESULTS

Life Cycle Inventory. While the functional unit is based on the management of 850 tonnes of mixed textile waste, it also represents the production of 280 tonnes of cellulosic fiber and 350 tonnes of polyester. Thus, there is a loss of 35% of the original mass. The majority of this (19% of the original mass) is associated with the initial ripping and shredding processes, where nonrecyclable components of the textile waste (e.g., zippers) are separated from the main stream. Other losses occur during the three filtration steps between the hydrolysis and fiber production processes.

The overall, principal material, and energy demands of the BRW scenario (Figure 2) are shown in Table 1. More detailed information by unit operation is provided in the Supporting Information.

Life Cycle Impact Assessment. The results of the LCIA are shown in Figures 4–10. The four columnar sets of data present benchmark scenarios for single-use of a forest cellulosic fiber (based on viscose), single-use of cotton, the BRW scenario, and that scenario but without ethylene glycol recycling ("BRW ex EG"). The results for each scenario are shown on the basis of the management of 850 tonnes of mixed waste, in accordance with the functional unit. Note also that "New PES synthesis" refers to the production of new polyester from the recycled terephthalic acid and ethylene glycol. In each figure, the error bars show the influence of optimiztic and

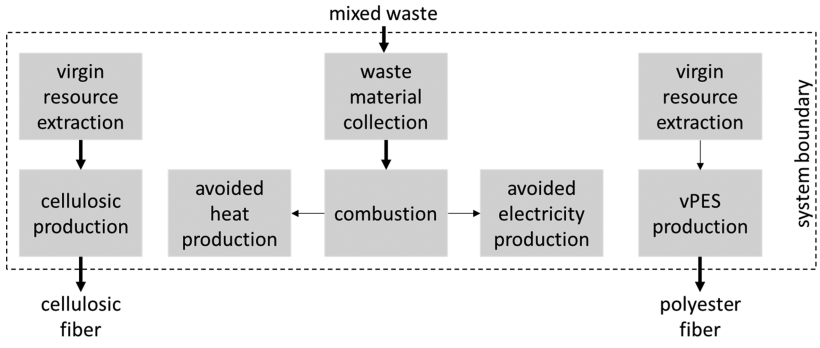

Figure 4. System diagram for single-use scenarios (thicker arrows indicate solid phase flows; vPES = virgin polyester).

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Table 1. Material and Energy Demands for BRW Process

flow	quantity per functional unit	denominating unit
waste textiles	850	tonnes
-waste cotton	383	tonnes
-waste polyester	467	tonnes
(new) ethylene glycol	8.08	tonnes
sodium hydroxide	547	tonnes
acetic acid	363	tonnes
sulfuric acid	513	tonnes
fresh water	1240	tonnes
electricity	0.61	GWh
natural gas	2.6	GWh

pessimistic assumptions about the emissions and resource use associated with the viscose or cotton production processes as described elsewhere in this paper (summarized in Sensitivity and Uncertainty Analysis and detailed in section 9 of the Supporting Information). The reader should bear in mind that the single-use scenarios have a long history of process optimization which BRW does not, so the fact that these results are of a similar scale is promising. Overall, the results suggest that the BRW process is competitive with the alternatives and that optimal environmental performance depends on the operation of the viscose facility.

The acidification potential results shown in Figure 5 are dominated by sulfurous emissions during the cellulosic fiber production processes, while the process for the acidification of the liquid stream also plays an important role in the case of the BRW scenario. The single-use cotton scenario also shows high acidification potential on account of ammonia emissions due to the use of nitrogen fertilizers, which is an issue of some concern in cotton cultivation (e.g., Tian et al).[19] There is relatively little benefit in avoiding the glycol recycling and separation process (compare the "BRW case" with "BRW ex EG") as the saved emissions are made up for by the emissions from additional (new) glycol production.

There is greater differentiation among the climate change results as shown in Figure 6. Cellulosic fiber production still plays a large role but the ethylene glycol separation process harms the overall competitiveness of the BRW process, as does the production of sodium hydroxide for the hydrolysis process. Emissions of the greenhouse gas carbon dioxide during polyester production are the main problem in the single-use cotton scenario and also play an important role in the single-use viscose scenario. Note that the primary energy consumption results closely resemble the results for climate change, so a figure for primary energy consumption has been omitted for brevity.

Among the freshwater ecotoxicity results in Figure 7, the single-use cotton scenario is the worst performer but within the uncertainty margins of the other scenarios. The primary driver of toxicity in this scenario is the emission to freshwater of herbicides and pesticides within the cotton agricultural sector, including dinitroaniline derivatives and a range of halogenated products. For the other scenarios, heavy metal emissions to air and freshwater from the viscose production process are the main contributors. Polyester synthesis also plays a role for the single-use scenarios. The overall profile of the results is similar for eutrophication potential (see Figure 8) though the outcomes for the different scenarios are closer to each other and polyester production plays a very small role. The main causes of eutrophication are phosphate emissions during cotton cultivation and viscose production.

Heavy metal emissions associated with the cellulosic fiber production processes, polyester synthesis, and acidification processes are the key determinants of the human toxicity potential (cancer) results in Figure 9. As the LCI database information is aggregated (cradle-to-gate) for the first two of these processes, it is unclear whether it is the inputs or the foreground processes that cause these emissions. In the case of the third process, it is the production process for the sulfuric acid that is the main contributor to these emissions. In the case of this indicator, recycling the ethylene glycol in the BRW scenario provides a small benefit compared with the production of virgin ethylene glycol. However, although both BRW scenarios perform better than the single-use scenarios, the overall difference is within the range of uncertainty

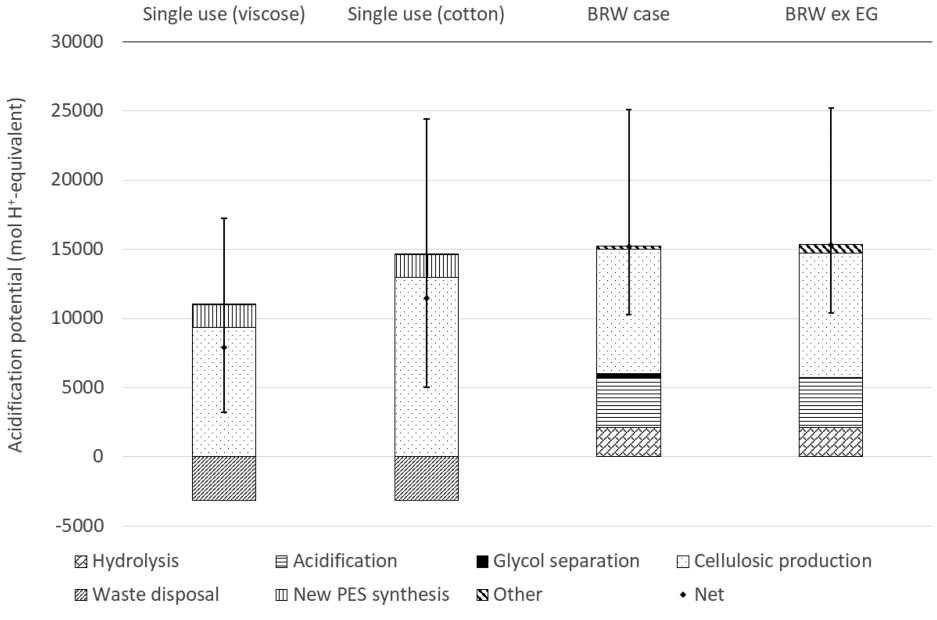

Figure 5. Acidification potential per functional unit, four scenarios.

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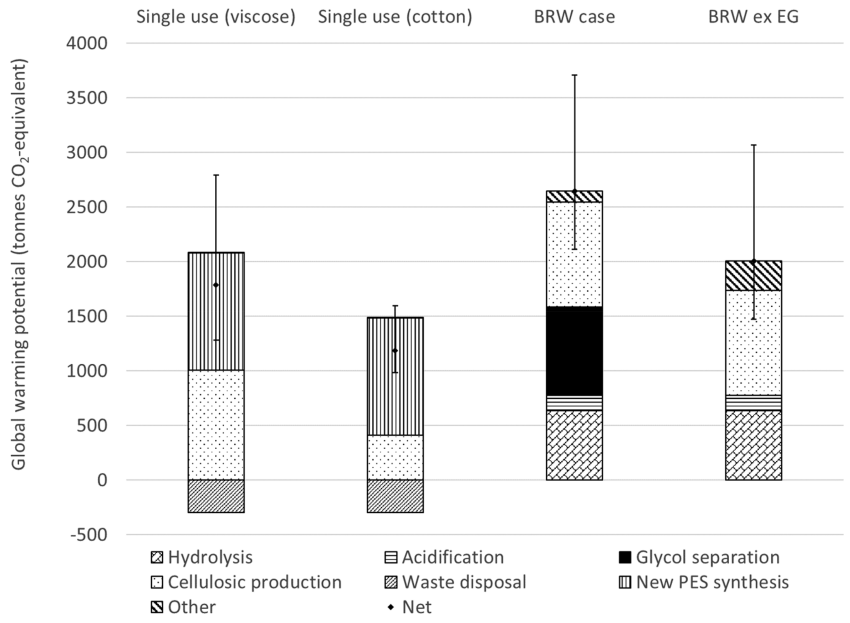

Figure 6. Contribution to climate change per functional unit, four scenarios.

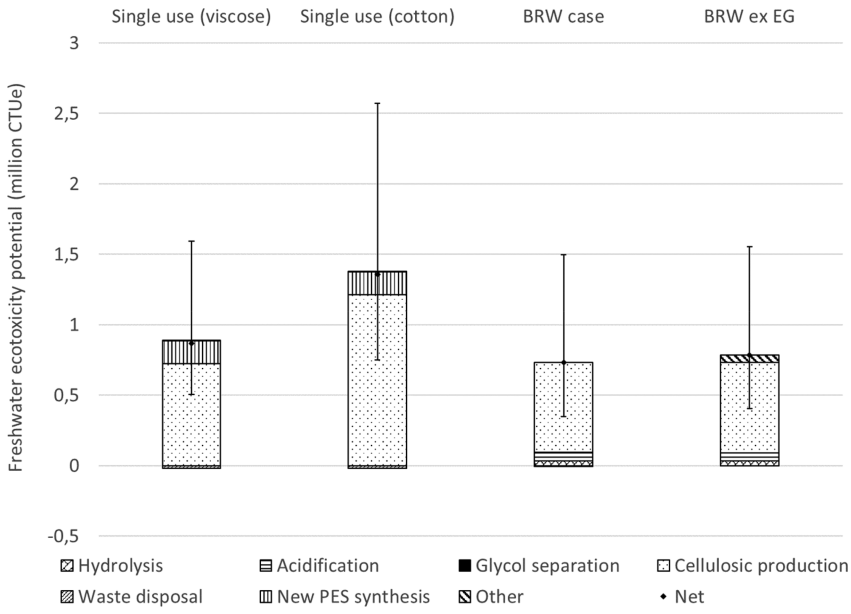

Figure 7. Freshwater ecotoxicity per functional unit, four scenarios.

associated with the selection of the cellulosic fiber production processes.

The results for noncancer human toxicity potential (Figure 10) are dominated by the atmospheric emissions associated with the viscose production process and upstream inputs to it (e.g., chemical production). In this case, there is apparently a significant difference between the single-use cotton scenario and the other three scenarios. The same can be said for the water scarcity results in Figure 11, except in this case the opposite outcome occurs: cotton production is water intensive and uses water in locations where water demand over availability is high. In comparison, viscose production from trees or recycled material uses little water if located in Sweden, where water demand over availability is low in most hydraulic catchments. Nevertheless, achieving the right concentration of sodium hydroxide in the hydrolysis step does require enough water to give the recycling scenarios a poorer performance against this indicator than the single-use viscose scenario.

DISCUSSION

With the exception of the (noncancer) human toxicity potential and the water scarcity indicators, the results for the four scenarios are within the range of environmental performance associated with the selection of the cellulosic fiber production processes. In other words, whether the recycling system is environmentally beneficial or not in relation to a conventional (waste management and production) system will for most indicators be determined by the environmental performance of the viscose or cotton production process, not the question of whether the raw material for fiber production is recycled or new. So according to the analysis presented here, the BRW process appears environmentally competitive with

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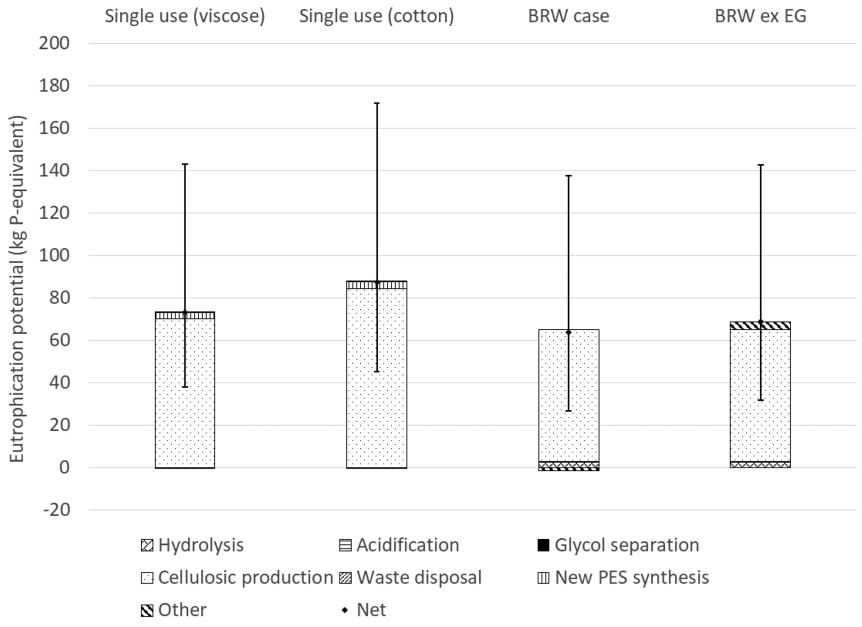

Figure 8. Eutrophication potential per functional unit, four scenarios.

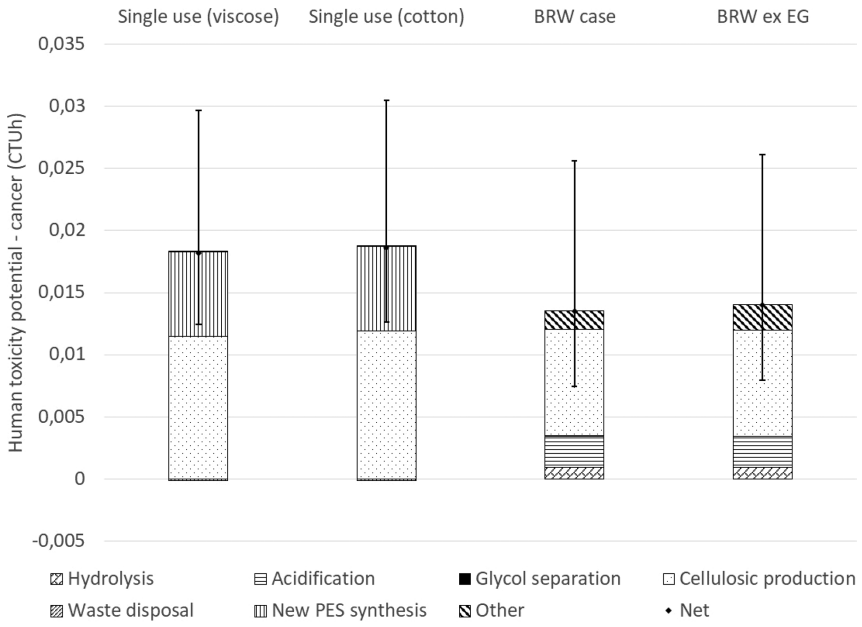

Figure 9. Human toxicity potential, cancer, per functional unit, four scenarios.

business-as-usual. This overall outcome is consistent with a recent LCA of another process for cotton recycling, which used scaled-up laboratory data to show significant water savings for the recycling process compared with cotton but climate impact similar to virgin viscose fiber production.[20] The outcome is also rather consistent with those of Östlund et al.,[10] who assessed a range of hypothetical scenarios for chemical (and mechanical) recycling of cotton/polyester blends and found that recycling can (compared to combustion with energy recovery) reduce or slightly increase climate impact, whereas substantial reductions in ecotoxicity are possible if cotton production is replaced. As in the present study, the climate benefits identified by Östlund et al.[10] depend primarily on assumptions about the environmental performance of fiber production processes (including the production of the recycled as well as the replaced fibers). One aspect to bear in mind regarding the performance of the single-use benchmarks is that they are based on combustion of waste in Sweden. In other jurisdictions where such waste is landfilled, the absence of energy recovery and the generation of methane from decomposing cotton can be expected to increase the climate impacts of the single-use benchmarks relative to the BRW system. This is likely to hold even where attempts are made to trap landfill gas and burn it for energy, as the energy recovery rate of textile combustion is superior.[21] As previously mentioned, it should also be borne in mind that this study compares industrial and agricultural processes (the single-use scenarios) which have been subject to worker-centuries of process optimization with a process (BRW) that only exists at laboratory scale. Therefore, it is to be expected that as work

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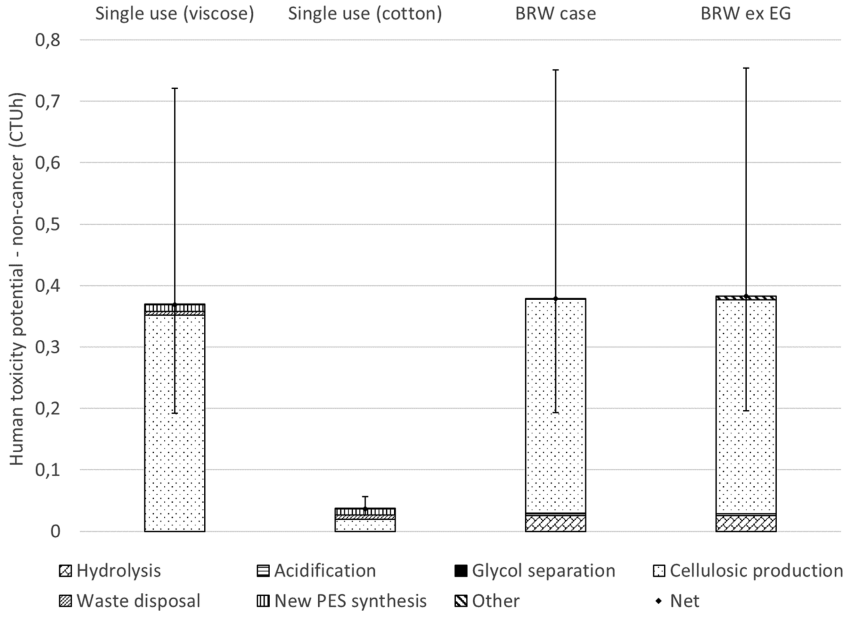

Figure 10. Human toxicity potential, noncancer, per functional unit, four scenarios.

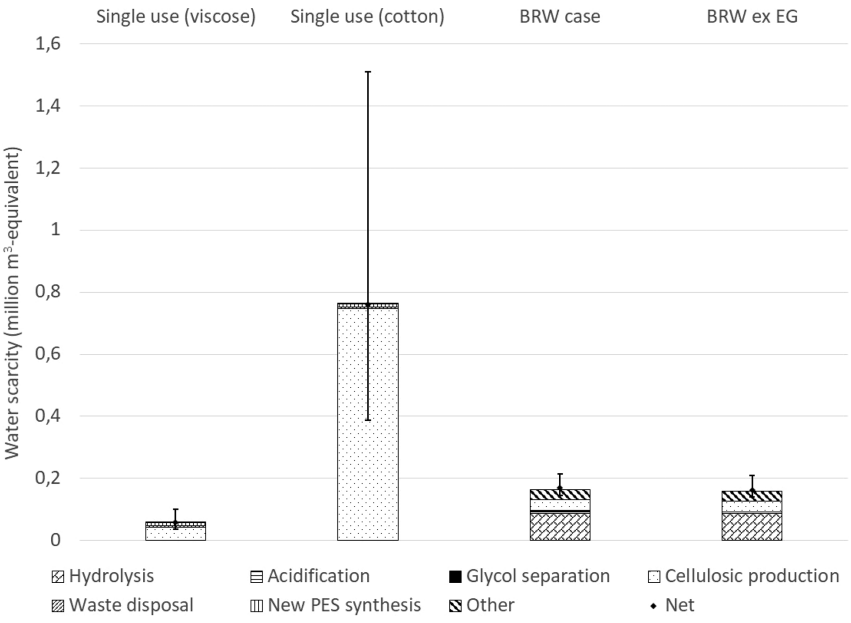

Figure 11. Water scarcity per functional unit, four scenarios.

proceeds on the development of this process, it is likely to become more efficient in its use of energy and materials.

With regard to improvement opportunities, another way of describing the outcome of this LCA is that process designers should focus primarily on selecting an optimal cellulosic fiber production process. However, since the other parts of the process have not undergone the same amount of process optimization, and their contributions to the results in several impact categories are considerable, process designers should also direct their attention on optimizing these other parts of the BRW process. For example, the model suggests that recycling the terephthalic acid is beneficial but recycling the ethylene glycol in the wastewater causes a higher climate impact because of the high salt content of the waste stream, which makes distillation a necessary unit operation. So in future experimental work it could be worthwhile to investigate the influence of varying the sodium hydroxide recycling rate on cost and environmental indicators with a view to optimizing the rate of salt generation.

The usual caveats regarding uncertainties in LCA apply here. First, this is a study of hypothetical, future production systems, based on the constraints and opportunities apparent at the present time. As such, there are some inherent uncertainties: (i) it is uncertain what technologies are relevant to study (thus the range of technologies assumed for production of regenerated cellulose fibers), how foreground systems will evolve from laboratory scale to full scale (assumptions had to be made regarding, for example, water use and chemical recycling), and how background systems will evolve (for example, electricity supply).[22] Furthermore, we relied on commercial LCI databases (Gabi Professional and Ecoinvent) for inventorying background systems and are therefore

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dependent on the quality of that data. We used LCIA models that are current norms in a European context but are subject to constant extension and renewal. Moreover, we used the currently dominating allocation practices for handling open-loop textile recycling,[6] but the recently issued circular footprint formula may challenge these practices and change the outcome of studies of open-loop recycling.[23,24]

As mentioned, other textile systems are feasible using different chemistries and different input materials. One example of an alternative method for handling mixed textile waste is the use of *N*-methylmorpholine N-oxide (NMMO) to dissolve the cellulosic material and leave the polyester in the solid phase. This was considered in a greenhouse gas life cycle assessment by Zamani et al.[4] and shown to be preferable to the default waste management method (combustion with energy recovery). Several subsequent papers have used data from that paper to build bigger system models (e.g., Dahlbo et al;[25] Fortuna and Diyamandoglu[26]), but there is relatively little data available for this or other mixed textile waste recycling processes. The present paper provides new model data which may be useful for other researchers looking for input to such larger system models. It will need to be borne in mind that the extension of this system to larger scales will require some means of producing recyclable material streams of suitable quality that do not exist today. This could mean better source separation and collection systems or better sorting systems for collected mixed wastes. It will be important to design such systems with the overall goal of minimizing their impacts, for example those associated with the logistics of collecting multiple separated textile streams.

CONCLUSIONS

This LCA suggests that alkaline hydrolysis of mixed textile waste is a promising direction for future research and development. While half of the indicators evaluated in the LCA favor the single-use scenarios over the recycling scenarios, in most cases the differences between the results calculated for the alternatives is less than the uncertainty associated with them. So overall, the recycling scenarios examined here performed about as well as the established single-use scenarios. Since the latter have been subject to process optimization over many years while the recycling process has only been demonstrated at laboratory scale, we may optimistically state that the relative performance of the recycling process can be expected to improve over time. Key aspects for future consideration include optimal selection of the regenerated fiber type; integrated regenerated fiber production facilities; the sodium hydroxide recycling rate; and the suppliers of chemical inputs to the process. Operating a plant at the scale described in this article will hopefully provide data enabling more precise evaluation of the environmental performance of the process and the know-how necessary to scale up to larger and more varied mixed textile waste streams.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acssuschemeng.9b01742.

Qualitative and quantitative details regarding the life cycle inventory of the recycling process in addition to basic data regarding the single-use processes (PDF)

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Notes

The authors declare no competing financial interest.

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