

Article

Comprehensive Meta-Analysis of Pathways to Increase Biogas Production in the Textile Industry

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Abstract: The textile industry is one of the largest environmental polluters in the world. Although waste management via anaerobic digestion (AD) is a sustainable strategy to transform waste into clean energy and water recovery, the efficiency of the AD process is reduced by the presence of recalcitrant materials, chemicals, and toxic contents. This study aims to investigate the performance of several chemical, physical, and biological pretreatments applied to improve the biodegradability of textile waste. We performed a meta-analysis with 117 data extracted from 13 published articles that evaluated the efficiency of pretreatments applied to textile waste prior to AD to increase biogas production measured as methane (CH₄) yield. Even though the majority of the studies have focused on the effect of chemical and physical pretreatments, our results showed that the application of biological pretreatments are more efficient and eco-friendlier. Biological pretreatments can increase CH₄ yield by up to 360% with lower environmental risk and lower operating costs, while producing clean energy and a cleaner waste stream. Biological pretreatments also avoid the addition of chemicals and favor the reuse of textile wastewater, decreasing the current demand for clean water and increasing resource circularity in the textile industry.

Keywords: textile residues; biotechnology; methane; circular economy; fibers; cotton



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1. Introduction

The textile industry is one of the largest polluting sectors worldwide, with an estimated waste production of 92 million tons per year [1], including pre- (i.e., agricultural production, fiber production, wastewater, solid waste) to post-consumer (i.e., manufacturing, logistics, retail and mixtures of discarded clothing or household items) waste in the supply chain [2]. Over 8000 chemicals (e.g., dyes, suspended solids, chlorinated aromatic hydrocarbons, surfactants, and heavy metals) are used in the textile supply chain [3,4]. As a result, effluents and solid waste with high loads of hazardous chemicals are discharged, thus increasing the toxicity of the produced waste, with a high pollution risk to the environment and human health [5].

Sustainable manufacturing is crucial to reducing the environmental impact of fashion and the textile industry. Projects and policies aiming at the sustainable development of the market, such as the [6], “Strategic Agenda on Textile Waste Management and Recycling”, Expert Network on Textile Recycling (ENTeR), and Conference of the Parties (COP 21), as well as the 2030 Agenda for Sustainable Development Goals (SDGs), have been important players in reframing textile production. Incentive actions for reuse are also crucial to the

implementation of the circular economy model as established by the European Union in this sector [7].

Approaches to minimizing textile waste and increasing life span such as clothing rental and repairing, the second-hand market, and reprocessing operations for the production of original or new products [7] are alternatives to manage the polluting potential. However, the advance of fast fashion follows a business model that produces large quantities of clothing and trends at low prices [1], and often of low quality. The low quality of textiles makes recycling unfeasible, restricting their use to the end of the waste management hierarchy—energy recovery—as determined in the in the Directive 2008/98/EC [8].

Anaerobic digestion (AD) is a widely applied biotechnology that has proven its effectiveness as a green solution for waste management, reducing the risk of contamination while producing energy as biogas. AD implementation in the textile industry can contribute to waste use as a resource for the generation of clean energy, as well as water reuse. However, the wide range of chemicals, organic pollutants, and recalcitrant compounds (i.e., polyacrylates, phosphonates, alkyl phenol ethoxylates, chloroform, heavy metals, and cotton-based recalcitrant material [9]) of textile waste poses challenges, reducing the efficiency of degradation if AD is applied as the only strategy [10].

Several studies have suggested that the application of pretreatments is advantageous not only to improve organic matter degradability, but also to increase the biogas yield [10–13] and further remove dyes and toxic compounds from wastewater and solid waste from aqueous solutions [4,10]. Pretreatments can be chemical, physical, biological, or combinations of these, and their performance is influenced by the chemical composition of the waste [14]. Physical pretreatments (i.e., thermal, mechanical, irradiation, ultrasound) act by disrupting cells through physical force [15]. As a result, the contact surface of the organic matter is increased by reducing the particle size, facilitating microbial attack [16]. Although physical pretreatments are advantageous since no toxic compounds are generated, some techniques (e.g., thermal) can increase energy costs, becoming unfeasible at a large-scale [16]. Chemical pretreatments (i.e., acid, alkali, organic solvents) act by breaking chemical bonds from complex structures, causing an internal increase in the surface by swelling of the cell [17]. Chemical techniques are highly efficient in degrading complex materials and are more often applied than biological and physical pretreatments [17]. However, they require extra care, since depending on the chemical reagents applied, toxic compounds can be formed. Some chemicals pretreatments such as sulphuric acid (H_2SO_4), hydrochloric acid (HCl), and nitric acid (HNO_3) can further damage operational equipment by corrosion and failure of metals [18–20]. Biological pretreatments (i.e., fungi, bacteria, microbial consortia, enzymes) act in synergy with microbial metabolism, promoting the acceleration of the degradation of organic matter [16]. Avoiding the addition and generation of harmful chemicals promotes an environmentally healthier AD system. In addition, the lower capital and energy costs compared to physical and chemical pretreatments make the application of biological techniques extremely attractive even at full-scale [19,21].

In this context, we performed a systematic review followed by a meta-analysis of the available data in the literature to evaluate the effect of pretreatments applied to textile industry waste. The aim of this study was to assess the efficiency of several pretreatments applied to textile waste prior to AD. Based on the peer-reviewed literature, we compared several physical, chemical, and biological pretreatments, as well as combinations of these, to identify the best choice in terms of the highest organic waste reduction through biogas production with the highest cost–benefit ratio.

2. Materials and Methods

The Preferred Reporting Items for Systematic Reviews and Meta-analyses (PRISMA) was used as a guide to conduct the execution of the sampling design, determining the search strategy, keywords, and exclusion/inclusion criteria for evaluating the quality of articles [22]. A systematic review followed by a meta-analysis was carried out using the Web of Science and Scopus databases and the Google Scholar search engine. We

filtered our search using keywords combined with the aid of Boolean operators (AND) and wildcards (*, \$): *anaerobic digestion*, *textile*, and *pretreatment*, from 1945 to 2021, English language, with article as the type of document. The eligibility criteria to include articles in the meta-analysis were: (i) textile waste as a unique substrate for AD; (ii) application of pretreatment for biogas production; (iii) measurement of methane yield from the organic fraction; (iv) experiments with specific descriptions and measurements for control and treatment samples.

Based on Hedges et al. [23], methane production data were quantified using the natural log response ratio (RR). Thus, the mean of the pretreatment performance (\bar{X}_T) was compared to the mean of the control (\bar{X}_C), following the equation

$$RR = \ln(\bar{X}_T / \bar{X}_C) \quad (1)$$

An unweighted meta-analysis was conducted to include the largest number of studies, even those that did not report a measure of variance for the response variable [24]. Mean effect sizes and 95% confidence intervals (CI) with bias correction were conducted using R software (R Core Team, 2021) using the “metafor” package (version 3.0-2). Pretreatments were considered significant ($p < 0.05$) when their mean effect and CI did not overlap the zero line. Mean and upper CI below the zero line indicate a negative response (i.e., treatment < control or treatment less efficient than control) while mean and lower CI above the zero line represent a positive response (i.e., treatment > control or treatment more efficient than control).

We assessed the potential production of CH₄ that could be generated from cotton and polyester waste if the most appropriate pretreatment is applied. Cotton and polyester waste data from the textile industry were collected, respectively, from [25,26]. The volatile solids (VS) and total solids (TS) content for each textile waste was based on data reported in the literature [27,28] (Equations (2) and (3); Table S1). The potential production of CH₄ was estimated based on the average methane yield of untreated and pretreated cotton and polyester, considering the pretreatments with the highest increase reported in this study (Table S2) and a lower calorific value of 9.97 kWh/m³ CH₄ [29] (Equation (4)).

$$GCW = (CW \times TS\%) \times OM_{\text{cotton}} \quad (2)$$

$$GPW = (PW \times TS\%) \times OM_{\text{polyester}} \quad (3)$$

$$CH_4 \text{ potential} = GPW \times CH_4 \text{ yield} \times 9.97 \quad (4)$$

where GCW is global cotton waste (million t VS/y), CW is the cotton waste (million t/y), GPW is global polyester waste (million t VS/y), PW is the polyester waste (million t/y), OM is the organic matter content in the waste (e.g., cotton, polyester) (VS%), and 9.97 is the lower calorific value of CH₄ (kWh/m³ CH₄).

3. Results and Discussion

A total of 117 data were extracted from 13 articles (Table S3) that applied pretreatments to improve AD of textile waste (Figure S1). The solid fraction of textile waste prevailed (81%) over the liquid fraction as the type of waste that undergoes pretreatment prior to AD. Wool and pure cotton were the waste sources with the largest contribution, ca. 38% each (Figure 1).

Cotton represents a robust fraction of textile waste [30]. Its chemical composition consisting mostly of cellulose, about 80% DW (Table 1), a polymer structure with high crystallinity that requires pretreatment for successful AD [31]. Wool is a natural protein fiber highly exploited as a raw material in the textile industry due to its qualities such as high heat retention, high stain and static resistance, and good flexibility [32]. As the highest methane yield is usually observed in protein-rich substrates [33], the high protein content in wool (70% DW) indicates its potentially high energy value (Table 1). In addition, wool and cotton come originally from the agricultural sector, where AD is largely applied as a “waste-to-energy” technology promoting waste recovery and energy generation [34].

Thus, both residues are strong candidates to be explored for energy recovery, as well as to strengthen the textile supply chain from its pre-consumer phase in agricultural production.

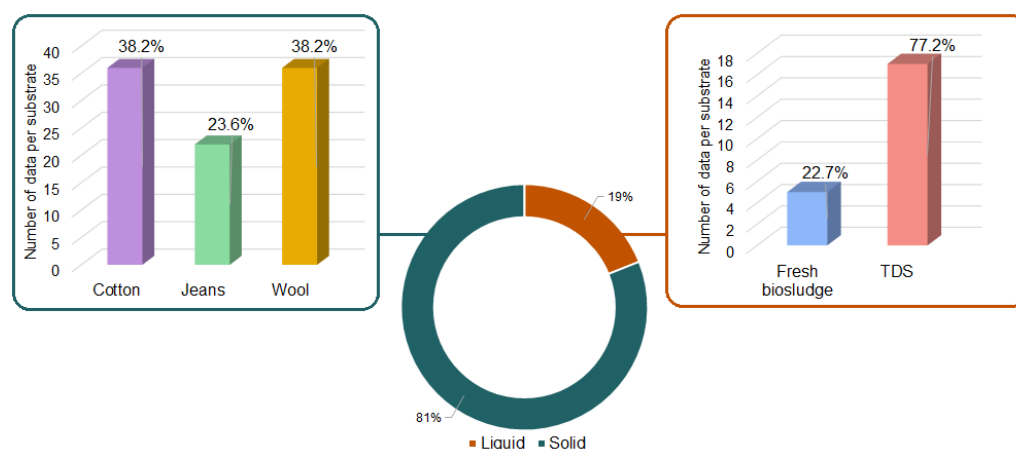


Figure 1. Different sources of textile waste used in AD reported in the literature included in the meta-analysis. TDS = textile dyeing sludge.

Table 1. Chemical characterization of different textile sources presented as averages ¹.

| | Cotton | Jeans | Wool | Fresh Biosludge | Textile Dye Sludge |
|---------------------------------------|--------|-------|--------|-----------------|---|
| TS (%) | 93.4 | 98.5 | 37.9 | 14.9 | 4.1 |
| VS (%TS) | 84.2 | 61 | 16.6 | 77 | 52.6 |
| pH | 7.1 | N.A. | 7.9 | 8.7 | 7.2 |
| Biological oxygen Demand (BOD) (mg/L) | N.A. | N.A. | N.A. | 887.3 | 1512 |
| Chemical oxygen demand (COD) (mg/L) | 2550 | N.A. | 2080.2 | 25,215 | 741.3 |
| Protein (%DW) | N.A. | N.A. | 70 | N.A. | N.A. |
| Heavy metals (mg/gSS) | N.A. | N.A. | N.A. | N.A. | Pb 0.04, Ni 0.10, Cd 0.03, Cu 0.26, Zn 0.29 |
| Cotton (%DW) | N.A. | 70 | N.A. | N.A. | N.A. |
| Polyester (%DW) | N.A. | 45 | N.A. | N.A. | N.A. |
| Cellulose (%DW) | 80 | N.A. | N.A. | N.A. | N.A. |

¹ References used are reported in Table S4. N.A. = not available

High consumer demand in the textile industry has driven several waste management strategies. Incineration and landfill are the main destinations of most textile waste at their end of life [1]. Although waste volume decrease by 90% is achieved via incineration and this process is viewed as an effective form of energy recovery [2,35], contaminated, damp, ripped, or stained textile waste does not contribute to energy recovery [36]. In addition, incineration can produce flue gases (e.g., sulphur dioxide (SO₂), HCl, hydrogen fluoride (HF), and nitrogen dioxide (NO₂)) that are harmful to the environment [35]. AD is, thus, the most promising technology when compared in terms of both environmental performance and energy recovery [37]. Even though it is a great challenge to stabilize the AD process since its stages are driven by microorganisms [38], the generation of clean energy and biofertilizer makes it advantageous from an environmental perspective [39].

Blue jeans waste mainly consists of cotton (70% DW) and polyester fiber (45% DW) (Table 1) and makes a large contribution to the solid fraction, representing 23% of all solid waste (Figure 1). Although the conversion of cotton to biogas through the breakdown of cellulose into simple sugars has been extensively described in the literature, its blending

with polyester is poorly described and requires further research [30,31,40]. Furthermore, as polyester represents a robust fraction of the textile sector and an increase in its production is projected due to cost-effectiveness in line with the consumption pattern of emerging countries, AD of this fraction requires attention [1]. The main challenge and bottleneck is the hydrolysis of polyester, as its large molecules hinder enzymatic attack by microorganisms, limiting the process to the surface of the material and, consequently, extending the duration of the process by months [7].

The liquid fraction of the residual organic matter represents 19% of all organic waste (Figure 1) and is mainly composed of textile dye sludge (TDS) (77.2%) and fresh sludge (22.7%) (Figure 1). Even though the liquid waste volume is much lower than the solid waste, the liquid fractions are highly toxic, with heavy metal content (Table 1), and their pollutant potential is very high [4], second only to tanneries and the pulp and paper industry [3]. High concentrations of chemical oxygen demand (COD) and biological oxygen demand (BOD), which are the analytical parameters of the degree of organic pollutants, are observed in the liquid fraction of textile waste, in the range of 741–25,215 mg/L and 887–1512 mg/L, respectively (Table 1). The amount and chemical content of textile effluents are of huge concern due to the presence of highly water-soluble chemicals. Conventional activated sludge systems or municipal sewage treatment systems are inefficient for the treatment of this residue [41], indicating a clear need for investments in water reuse and energy recovery from this waste stream. Successful pretreatment application to textile effluents has led to dye removal, decolorization of wastewater, and reduction of toxicity [4,10]. Moreover, the proper selection of the AD pretreatment results in a cost-effective and secondary treatment to alleviate environmental damage [4]. Pretreatments promote the reduction of pollutant loading, mitigating impacts on environmental and human health. Furthermore, water reuse can supply the high water demand of the textile industry itself.

Several pretreatments such as physical, chemical, biological, and combination (Figure 2, Table S2) treatments have been applied to improve AD. Pretreatments applied alone, especially physical and chemical, are the most explored, while their combinations have been less examined (Figure 2, Table S2). This could be explained by the high capital cost investment required to integrate pretreatments into the AD process, and also by the difficulty of large-scale implementation [40].

Heat application (i.e., autoclave and thermal) is the most reported treatment among physical pretreatments, alone and in combination (Figure 2). High temperatures are successful in lysing cells and increasing the solubilization of organic materials such as polysaccharides and protein, increasing soluble chemical oxygen demand in the liquid and solid fraction of the textile waste [42,43] and favoring biogas generation.

Sonication and liquid nitrogen (LN₂) are mechanical pretreatments that are highly efficient in the disruption and disintegration of complex bonds in the substrate chemical structure [44,45]. Although the application of these pretreatments has a positive effect, the increase in methane yield is limited, which may be a result of the reduction and/or inhibition of AD caused by the presence of some chemicals released from the substrate. As a result, the implementation of these pretreatments may become economically unfeasible [45].

The diversity in chemical pretreatments is the largest (Figure 2), with microaeration ($n = 11$) being the most applied. Oxygen addition impacts the biological microbial process and microbial electrolysis in addition to inhibiting the production of hydrogen sulfide (H₂S), promoting a more stable AD process [27]. Other chemical pretreatments such as alkaline, acids, and organic solvent additions have been shown to be highly efficient in breaking down complex structures and increasing the availability of fermentative sugars for enzymatic hydrolysis [46]. However, when applied at high doses, they can generate methanogenesis-inhibiting bioproducts such as furfural and vanillin [47].

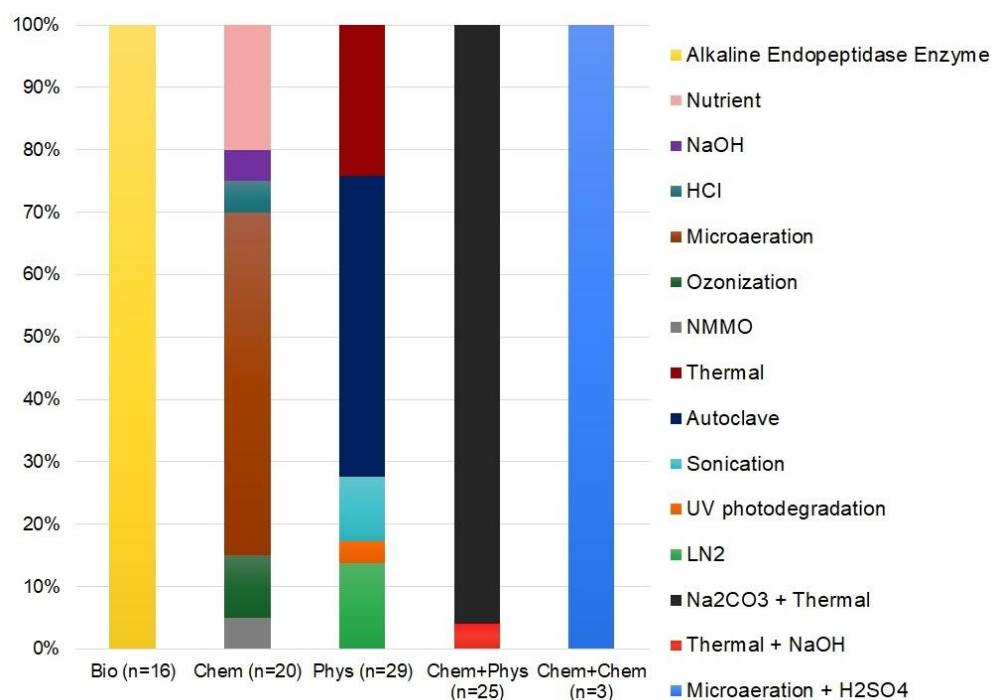


Figure 2. Diversity of methods composing the pretreatment categories applied to improve AD of textile waste reported in articles included in the meta-analysis. *Biological:* alkaline endopeptidase ($n = 16$); *chemical:* nutrient ($n = 4$), NaOH ($n = 1$), HCl ($n = 1$), microaeration ($n = 11$), ozonization ($n = 2$), N-methylmorpholine N-oxide (NMMO) ($n = 1$); *physical:* thermal ($n = 7$), autoclave ($n = 14$), sonication ($n = 3$), UV photodegradation ($n = 1$), liquid nitrogen (LN₂) ($n = 4$); *chemical + physical:* Na₂CO₃ + thermal ($n = 24$), thermal + NaOH ($n = 1$); *chemical + chemical:* microaeration + H₂SO₄ ($n = 3$).

Heat combined with sodium hydroxide (NaOH, $n = 1$) and sodium carbonate (Na₂CO₃, $n = 24$) were the thermochemical pretreatments reported (Figure 2). The predominance of Na₂CO₃ + thermal can be a consequence given the negative effects of sodium hydroxide such as corrosion, need for neutralization, and generation of hazardous content to the environment [31]. In contrast, sodium carbonate is successful in reducing cellulose crystallinity and consequently achieves high bioconversion of lignocellulosic content in addition to low cost [31].

As the use of enzymes provides a very selective and specific action on the organic matter [48], the predictability of its response could suggest a lower need for extensive testing, as required in other pretreatments. Therefore, the commercial enzyme alkaline endopeptidase ($n = 16$) was the only biological pretreatment applied to wastes from the textile industry (Figure 2, Table S2).

Biological pretreatments are the most eco-friendly, as they have a relatively low energy cost and require no addition of chemicals/inhibitory compounds. Thus, they generate less pollution and high methane yields [49]. The application of alkaline endopeptidase led to an increase in methane yield by up to 360% (RR= 1.28, 95% CI: 0.15 to 2.41, $p < 0.05$), indicating high efficiency as a pretreatment for the textile industry (Figure 3, Table S2). The use of commercial enzymes has become more attractive due to their pure nature of speeding reactions, thereby making biodegradation faster [50]. Addition of the alkaline endopeptidase enzyme shows stable performance with high protein solubilization, which improves methane production [42,50]. In fact, the success of enzyme application has led to its impressive growth in the last decade in the textile sector [48]. However, the low number of enzymes thus far investigated highlights the crucial need for more research in the area for the development of new enzymes with higher efficiencies targeting textile wastes.

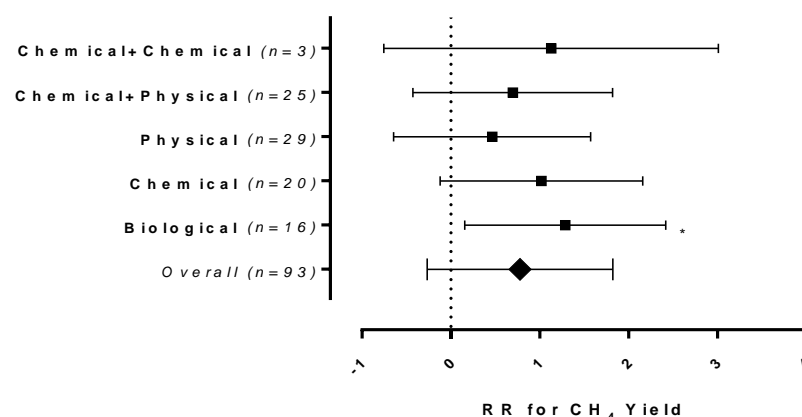


Figure 3. Effect size by natural log response ratio (RR) of methane yield with 95% confidence interval (CI) (p -value = 0.05), comparing the performance of the biological, chemical, physical, chemical + physical, and chemical + chemical pretreatments. Significant code $p \leq 0.05$ (*); n = number of effect sizes per treatment type.

Despite the superior performance of biological pretreatments (Figure 3), physical and chemical pretreatments remain the most applied on textile waste (Figure 2). This is probably due to their high performance when applied on other waste sources. However, our result (Figure 3) clearly shows that their application may not necessarily pay off and can even lead to decreases in methane yield in comparison to untreated controls, especially when applied to cotton waste (Table S2). Therefore, pretreatments should be carefully chosen before large-scale application.

The use of inappropriate pretreatments can, in fact, reduce the methane yield [51]. For instance, formation of inhibitory/toxic compounds following pretreatments, such as volatile fatty acids (VFAs) [52,53], can cause negative effects on biogas production due to the low capacity to degrade specific organic compounds and to deal with usual chemical loads in textiles such as dyes.

Chemical pretreatments used both individually and/or in combination showed a large variability over methane yield (Figure 3). This can be explained by the choice of additive (i.e., H_2SO_4 , $NaOH$) and the concentration added, which requires care, since high doses can inhibit methanogenesis [39,47].

Even though significant results were obtained in this study, our analysis must be viewed with caution, as the application of pretreatments in textile waste is limited. Despite biological pretreatment having the highest performance (Figure 3), the same efficiency cannot be assumed for all textile wastes, as the prevailing chemical composition of the substrate is likely to have an effect on its efficiency. Therefore, careful case-by-case attention is needed to the individual performance of the different substrates subjected to pretreatments (Table S2), especially if we consider textile residues with strong environmental impacts such as cotton and polyester [31].

In fact, some textile wastes, e.g., textile dyeing sludge (TDS), have high loads of dyes, auxiliary chemicals, surfactants, and heavy metals [54]. Such chemicals can severely impair the AD microbial community, especially during hydrolysis and acidogenesis [53]. AD alone has shown CH_4 yield of 0.1 mL/gVS, which is dramatically increased after its combination with pretreatment to 56.1 mL/gVS, corresponding to an increase of 56,000% (Table S2).

Cotton and polyester represent 37% and 63% of the total fibers that are produced within the textile industry, respectively [55]. High energy demand is required for the production of those fibers and their transport to their final destination. Energy consumption is estimated at 48 kWh and 101 kWh just to produce one kilogram of cotton or polyester fiber, respectively (Figure 4A).

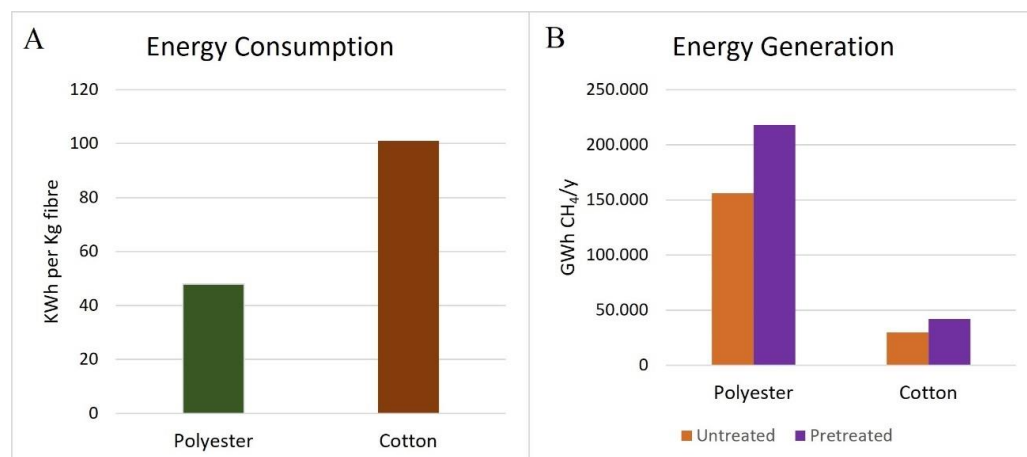


Figure 4. Consumption and generation of energy from the main textile fibers (cotton and polyester). (A) Energy consumption per kilogram of textile fiber produced, adapted from [1]. (B) Global energy generation potential from polyester and cotton untreated (orange) and pretreated (purple).

Replacing virgin cotton and polyester with second-hand clothes can save up to 65 kWh and 90 kWh of energy per kilogram of fiber reused, respectively [7]. However, the lifespan of textile products is continuously decreasing [7] and, therefore, the reuse of clothing as a strategy to reduce energy consumption is inefficient.

The destination of textile waste for AD can be a sustainable alternative for more efficient energy recovery and supply to the production chain. Considering the global annual waste production of cotton (11.7 million tons) and polyester (42 million tons) generated by the textile industry alone [25,26], we estimated a potential energy production of 155.835 GWh and 298.805 GWh from polyester and cotton, respectively (Figure 4B), which can be increased by applying the pretreatment with the best performance for those types of residues (Table S2).

4. Conclusions

Our results showed that biological pretreatments, of pretreatments commonly applied in the textile industry, promote larger biogas production via AD. In comparison with chemical and physical pretreatments, enzyme pretreatment may lead to an increase in methane yield up to 360% on average. Biological pretreatments also require no addition of chemicals and favor the reuse of textile wastewater, decreasing the current demand for clean water while increasing resource circularity in the textile industry. Moreover, biological pretreatments are very efficient in removing highly soluble chemicals that cause damage to human and environmental health, and given their lower energy demand, the cost-benefit ratio is expected to be low. The implementation of AD improved by pretreatment in the textile industry promotes adequate management of the huge amount of waste generated, converting textile residues into economic value for the industry in line with the circular economy. However, more research is crucial in the quest for new enzymes with higher efficiencies, as well as extensive screening of largely neglected residues, e.g., textile wastewater, in order to bring this sector to a more sustainable model.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/en15155574/s1>, Figure S1. Flow diagram summarizing quantitatively the selection of studies from the systematic review following the Preferred Reporting Items for Systematic Reviews and Meta-analyses (PRISMA, <http://www.prisma-statement.org/>). *n* = number of articles; Table S1. Global production of fiber waste from the textile industry and volatile solids (VS) and total solids (TS) content; Table S2. Summary of pretreatments applied to different sources of textile waste reported in the systematic review; Table S3: Studies included in the meta-analysis.

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