

## Article

# Environmental Profile Study of Ozone Decolorization of Reactive Dyed Cotton Textiles by Utilizing Life Cycle Assessment

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**Abstract:** Research approaches on the use of ecotechnologies like ozone assisted processes for the decolorization of textiles are being explored as against the conventional alkaline reductive process for the color stripping of the cotton textiles. The evaluation of these ecotechnologies must be performed to assess the environmental impacts. Partial “gate to gate” Life Cycle Assessment (LCA) was implemented to study the ozone based decolorization process of the reactive dyed cotton textiles. Experiments were performed to determine input and output data flows for decolorization treatment of reactive dyed cotton textile using the ozonation process. The functional unit was defined as “treatment of 40 g of reactive dyed cotton fabric to achieve more than 94% color stripping”. Generic and specific data bases were also used to determine flows, and International Life Cycle Data system (ILCD) method was selected to convert all flows into environmental impacts. The impact category “Water resource depletion” is the highest for all the ozonation processes as it has the greatest relative value after normalization amongst all the impact indicators. Electricity and Oxygen formation were found to be the major contributors to the environmental impacts. New experimental conditions have been studied to optimize the impacts.

**Keywords:** life cycle assessment; normalization method; environmental impacts; ozonation process; decolorization; reactive dyed cotton textiles; “gate-to-gate” life cycle assessment (LCA)



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## 1. Introduction

Reactive dyes constitute nearly 50% of the worldwide market for the coloring of cellulose-based fibers [1]. However, the coloration industry faces some general problems such as faulty or uneven dyeing and the presence of color patches on the surface of the textile fabrics during coloration and subsequent processing operations [2,3].

To rectify these problems, the normal approach practiced in the coloration industry is destructive stripping. But, this traditional and commonly employed technique consists of huge amounts of various oxidizing and/or reducing agents in a single color stripping process, such as hydrogen peroxide, sodium hypochlorite, chlorine dioxide, and the dichromate salts, as well as thiourea dioxide, sodium hydrosulphite, and decroline [4]. In addition to this, these traditional color stripping methods involve high temperature and use of chemicals, both of which contribute to ecological pollution loads, huge liquor consumptions, and high production costs [5,6]. Therefore, a sustainable process needs to

be implemented to overcome these drawbacks. Previous studies demonstrated the use of the biological stripping method as an ecofriendly and cost effective approach [4,7]. Recent studies showed that the photocatalytic system with UV/ $\text{Na}_2\text{S}_2\text{O}_4$ , used for color stripping, was more energy effective, ecological, and a sustainable alternative [8]. In our study, we have proposed the use of the ozonation process for the color stripping of the reactive dyed fabric in a pilot scale process. No previous studies are available on the environmental impact assessment of the designed color stripping process.

A significant contribution to environmental pollution and resource utilization has been caused by the textile industry [9]. Therefore, the textile industry (TI) is exploring emerging and cleaner technologies in order to minimize the use of natural resources. Further, the TI seeks to continuously improving sustainable activity techniques, thus aiming for zero emissions [10]. In that context, it is important to reduce the amount of textile waste due to manufacturing defects without increasing the overall impact.

The life cycle assessment (LCA) is defined as a compilation and assessment of the inlets, outlets, and potential environmental impacts of a process or product through its life cycle. It is a vital tool to carry out the environmental analysis [11]. LCA is a model to determine the manufacturing methods either they are sustainable or cyclic production and find a substitute ecofriendly production process. LCA studies are principally determined as “gate-to-gate” or “cradle to grave” [12].

For the color stripping process, the environmental impacts of the proposed ozonation method has to be quantified in order to justify the profile of the process [13]. In our study, we used gate-to-gate LCA methodology. The detailed color stripping and mechanical property characterization has already been discussed in a previous paper [14]. The environmental assessment in our work is based on defining a functional unit color stripping of 40 g of reactive dyed cotton fabric to achieve color removal, and the determination of different environmental impact categories for the ozone assisted color stripping method.

The aim of the study was to highlight the main contributors to the environmental impact of the ozone stripping process and then to find the best conditions for reactive dyed decoloration. In addition, this work intends to identify and evaluate the potential impact of the ozonation process used and also encourages the sustainability profile of the process.

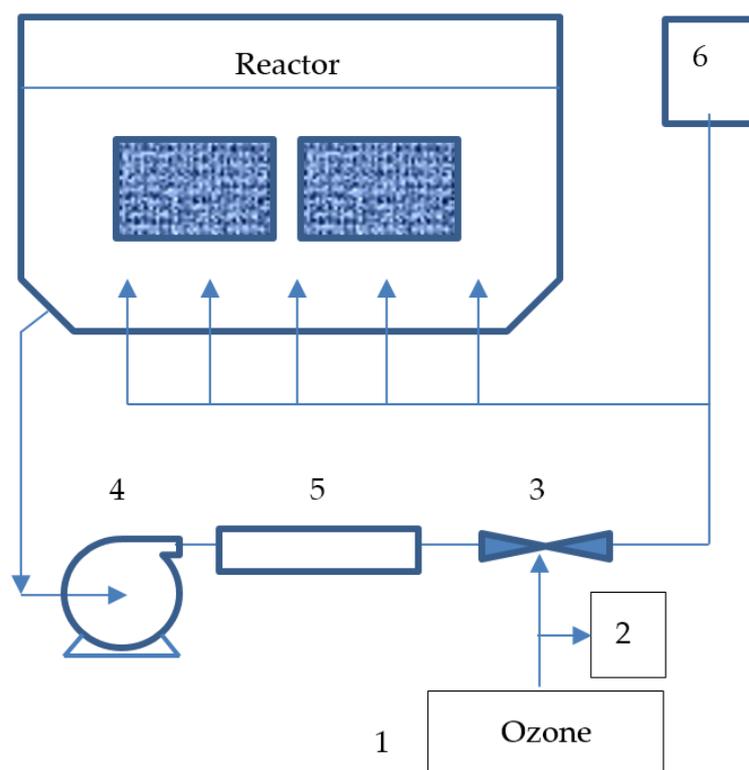
## 2. Experimental Work

### 2.1. Woven Cotton Textile

A 100% Cellulosic (cotton) woven fabric ( $150 \text{ g/m}^2$ ) was implemented in this study. The cotton fabric was dyed with a 1% reactive dye (C.I. Reactive Black 5). This dyed cotton fabric was used for the decolorization treatment of the fabric.

### 2.2. System Considered: Color Stripping Using the Ozone-Assisted Treatment

The ozone-assisted process was carried out using a pilot scale ozone reactor at the Unilasalle laboratory, France. The ozonation system is described in Figure 1 [14].



**Figure 1.** Pilot ozonation (1, ozone generator; 2, analyser ozone; 3, venturi injection system; 4, circulation pump; 5, filter; 6, dissolved ozone analyzer and pH meter) [14].

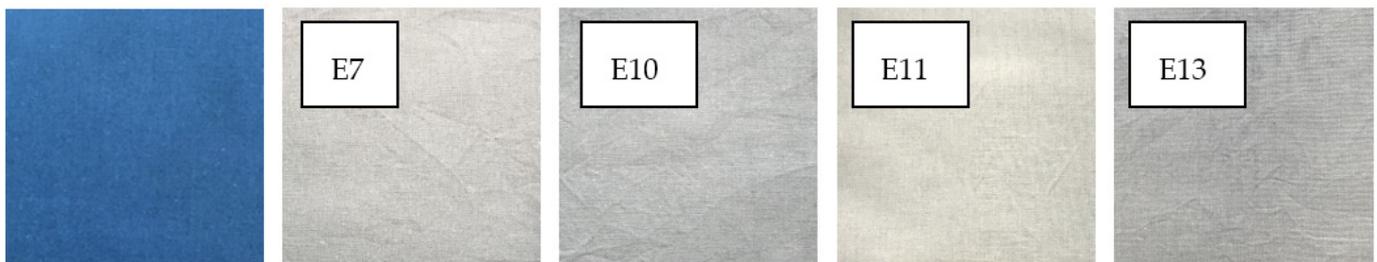
Ozone is produced by the electric discharge in oxygen provided by liquid oxygen pressurized bottles. Ozone transfer from the gas phase to the liquid phase is an important process to obtain the dissolved ozone in water in the reactor. Various techniques of the gas dispersion are applied in practice and diffusers, static mixer, injection etc. are the most popular ones [15]. In this study, we used the venturi injection process.

The oxygen  $O_2$  flow rate is constant at  $F = 0.3 \text{ m}^3/\text{h}$ , and the amount of ozone is measured in situ. The excess ozone is then destroyed in a 0.8 kW ozone destructor ODT-003.

The water bath used was at a fixed volume of 60 L of tap water. The circulation pump of the reactor has a power of 0.75 kW, and we made the assumption that only 10% of the power is required. All the experiments are made at room temperature. The pH value was regulated by adding phosphoric acid (PanReac AppliChem) and sodium hydroxide (EMPLURA<sup>®</sup> Merck, Germany). The pH was measured in situ during the ozonation process.

A Box Behnken experimental design matrix was setup to find the best experimental conditions for the decolorization with varying ozone concentration, pH, and treatment time, as described in our previous paper [14].

The 40 g blue dyed cotton fabric was placed in the reactor and subjected to the ozone treatment. As a result, the treated fabric started to decolorize and the color stripping % was measured using a spectrophotometer (Figure 2).



**Figure 2.** Cotton sample before and after ozonation treatment [14].

In this paper, we only considered experiments with stripping values more than 94% (Table 1). The best stripping was obtained with experiment E11 performed at pH 5. It had an ozone concentration in the oxygen gas flow of  $85 \text{ g/m}^3$  NTP (normal pressure and temperature) and a treatment time of 50 min. We considered the E11 experiment as a reference. In the experiments E7 and E8, the treatment time decreased to 30 min only, while a lower ozone concentration was used in E10 and E12. The E13–E16 experiments all had less ozone and less time, yet the stripping results were not good as compared to the reference.

**Table 1.** Ozonation experimental conditions with color stripping %.

Sr. No.	pH	Concentration Ozone	Time	Color Stripping
		( $\text{g/m}^3$ TPN)	(min)	%
E7	7	85	30	95.1
E8	3	85	30	97.45
E10	7	45	50	94.3
E11	5	85	50	97.6
E12	3	45	50	97.5
E13	5	45	30	94.6
E14	5	45	30	94.1
E15	5	45	30	94.65
E16	5	45	30	93.9

### 2.3. Material and Energy Requirement

The amount of resources required for the treatment of 40 g reactive dyed fabric was estimated from the treatment parameters and the characteristics of the devices of the process (Table 2).

**Table 2.** Ozone and energy requirements reference process E11.

Sr. No.	Inputs from Technosphere	Quantity
1	Energy for ozone generation with plasma treatment (kWh)	0.213
2	Energy for the circulation pump (kWh)	0.0625
3	Energy for the ozone destructor ODT-003 (kWh)	0.077
4	Oxygen (Kg)	0.357

#### a. Oxygen $\text{O}_2$ and ozone $\text{O}_3$ requirements:

The amount of  $\text{O}_2$  required was calculated from the flowrate and the treatment time. For the reference process E11, the treatment time was 50 minutes. So the amount of  $\text{O}_2$  required was  $0.25 \text{ m}^3$  corresponding to 0.357 kg of Oxygen as the oxygen ' $\text{O}_2$ ' density is  $1.429 \text{ kg/m}^3$ .

The concentration of  $\text{O}_3$  in the oxygen flow was constant. Thus, the total amount of  $\text{O}_3$  produced was calculated from the volume of oxygen ' $\text{O}_2$ ' gas used. With the oxygen concentration of  $0.85 \text{ g/m}^3$ , the  $\text{O}_3$  amount produced was equal to 21.25 g.

#### b. Energy requirements: Energy-associated concerns:

- Ozone generation with plasma treatment: Specific energy required to produce one kg of ozone from liquid oxygen was 7–13 kWh/kg O<sub>3</sub> [16]. An average value of 10 Wh/g O<sub>3</sub> was selected for our study, and thus this energy in the reference experiment was 212.5 Wh.
  - Ozone destructor ODT-003 operated at a power of 0.8 kW, which was associated with the maximum gas flow rate of 3.7 kg/h [16], or 2.59 m<sup>3</sup>/h with oxygen gas. As we used only 0.3 m<sup>3</sup>/h, then the power needed is 0.092 kW which when multiplied by the treatment time, yields the quantity of energy used. With experiment E11, which was carried for 50 min, the ozone destructor energy was 77 Wh.
  - Water circulation pump of the reactor: Multiplying the 0.075 kW power by the treatment time provided the energy used, and for the E11 experiment, it was 62.5 Wh.
  - For the reference treatment, E11, the total electricity requirement was 0.352 kWh.
- c. Chemicals

The water bath was made with tap water. In case of the reference process at pH = 5, the amount of phosphoric acid and sodium hydroxide used were 6.75 and 3.65 g, respectively.

### 3. Life Cycle Assessment

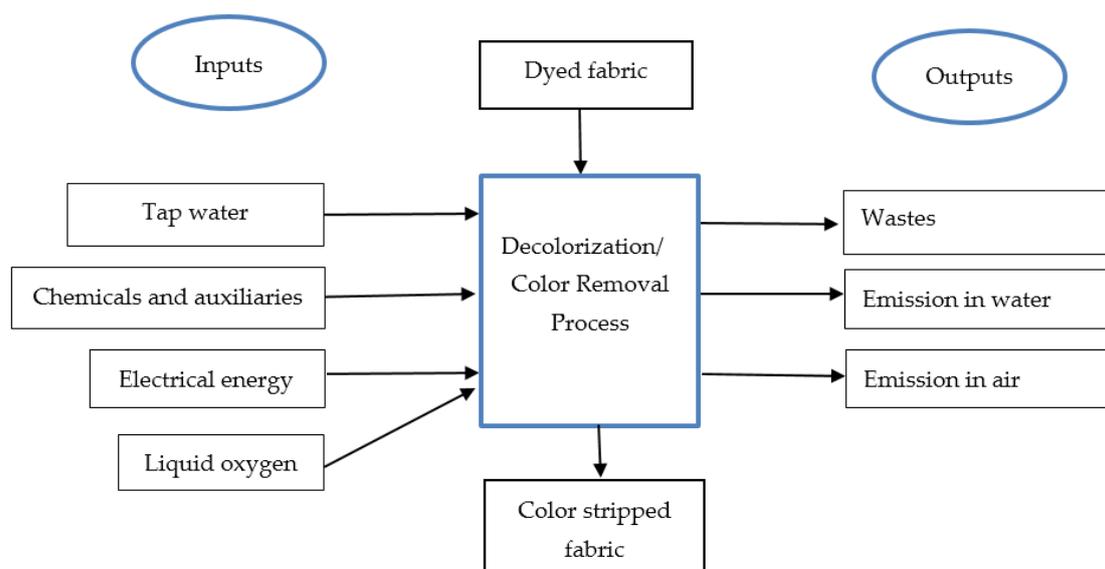
The LCA was modeled with the SIMAPRO LCA software tool as per the international standard. The decolorization or color stripping of textiles is a unit process carried out in the textile production value chain to rectify the faults or unevenness issues occurred during cotton textile manufacturing. “Gate-to-gate” LCA analysis considers only the color stripping process to study the environmental profile of the ozone-assisted process. The method used for the assessment of the environmental impacts was from International Reference Life Cycle Data System ILCD 2011 Midpoint+ V1.07/EU27 2010, equal weighting.

From the 16 impact categories of the ILCD method, the 6 following have been reported: climate change; water resource depletion; human toxicity (cancer effects); freshwater ecotoxicity; mineral, fossil, and renewable resource depletion; and the ionizing radiation of human health (HH).

To compare the significance of each impact category, they were all normalized using the 2010 normalization factors related to the EU-27 impacts [17]. In this study, the environmental impacts of a European person annually in 2010 are concerned.

#### 3.1. Goal and Scope Definition

The functional unit was defined as treating “40 g of dyed cotton fabric to achieve specific decolorization”. Various process scenarios leading to dyed fabric decolorization were studied for ozone-based color stripping processes. For the proposed ozone-based color stripping process, the “gate-to-gate” system boundaries considered the decolorization step for the manufacturing of chemicals and electrical energy (see Figure 3). Dyed woven cotton fabric manufacturing and chemical transportation were excluded. We hypothesized that the color stripping process was carried out in France. Production sites of chemicals and energy were in Europe. The following elements were outside the system boundaries: transport of chemicals and the fabrication/maintenance of the ozone machine and wastewater treatment. In this model, tap water was considered to minimize the impacts due to the use of deionized water or reverse osmosis water. There may be slight variations in the actual results due to the use of tap water. Moreover, the catalytic process was utilized for the ozone destruction, and hence the output was considered in terms of energy utilized for the destruction of the leftover ozone. Drying the samples was excluded from this study as it did not differ from one treatment to another.



**Figure 3.** System boundaries of the decolorization process of the reactive dyed cotton.

### 3.2. Life Cycle Inventory

For the ozone-assisted process, the experimental data was used considering the pilot scale designed machine. The scenarios were determined via laboratory experiments. From these scenarios, data were obtained to quantify flow inputs (consumed resources) and outputs (emissions or outcomes of the process). In our studies, data were obtained from several sources (Table 3). Specific data from experiments carried out in the laboratory and the production data was collected from the ECO INVENT database. These inventory data included the production of chemicals, liquid oxygen, and tap water in Europe (RER datasets), as well as electricity production and distribution in France (FR datasets).

**Table 3.** The life cycle inventory for the decolorization of 40 g of reactive dyed cotton fabric using the ozonation technique (reference the E11 experiment).

Inputs	Unit	Amount	Description	Source
Phosphoric acid	g	6.76	Phosphoric acid, industrial grade, without water, in 85% solution state {RER}   purification of wet-process phosphoric acid to industrial grade, product in 85% solution state   Alloc Rec, S	Eco-invent database
Tap water	mL	60,000	Tap water {Europe without Switzerland}   tap water production   underground water without treatment   Alloc Rec, S	Eco-invent database
Sodium hydroxide	g	3.55	Sodium hydroxide, without water, in 50% solution state {RER}   chlor-alkali electrolysis, diaphragm cell   Alloc Rec, S	Eco-invent database
Electricity	kWh	0.352	Electricity grid mix, AC, consumption mix, at consumer, 230 V FR, S	Eco-invent database
Oxygen	g	0.357	Oxygen, liquid {RER}   air separation, cryogenic   Alloc Rec, S	Eco-invent database
Outputs	Unit	Amount	Description	
Phosphoric acid	g	6.76	Wastewater content	
Tap water	mL	60,000	Wastewater content	
Sodium hydroxide	g	3.55	Wastewater content	

#### 4. LCA Results

##### 4.1. LCIA Results and Interpretation for the Reference Scenario

The main environmental impacts are described in Table 4. The total greenhouse gas (GHG) produced by the ozone treatment was 213 g of equivalent CO<sub>2</sub>. The water depletion was 168 L, while the resource depletion was 14 mg equivalent to Sb. The ionizing radiations were equivalent to 190 becquerel of the U235. The fresh water ecotoxicity was equivalent to 2 comparative toxic units (CTU), while the cancer human toxicity was calculated at  $0.02 \times 10^{-6}$  CTU. The normalization method was added to describe the extent to which the impact categories had a significant influence on the environment [18]. The normalized factor is the environmental impact caused annually by the activities of an average European, it is expressed as “person year equivalent”, PEq.

**Table 4.** Impact categories and normalized values for the impacts in the ozonation process E11.

Impact Category	Unit	Value (Unit: See Column)	Normalized Value (Unit: PEq.)
Climate change	kg CO <sub>2</sub> eq	0.21388189	0.0000235
Mineral, fossil, and renewable resource depletion	kg Sb eq	0.00001360	0.000135
Ionizing radiation HH	Kbq U <sub>235</sub> eq	0.19096354	0.000169
Freshwater ecotoxicity	CTUe	2.01311521	0.000229
Human toxicity, cancer effects	CTUh	0.00000002	0.000549
Water resource depletion	m <sup>3</sup> water eq	0.16857161	0.002073

The LCA normalized results for every impact category in the ozone reference process (E11) are displayed graphically in Figure 4, with the same equivalent person year unit. The four major impacts are as follows: water resource depletion, human toxicity, cancer effects, freshwater ecotoxicity, and the ionizing radiation HH. The main environmental impact for the reference process E11 concerned the water resource depletion, as it had the greatest relative value after normalization amongst all of the impact indicators. From Figure 4, we observed that there was a minor impact on climate change, as well as the mineral, fossil, and renewable energy depletion.

##### Interpretation

Considering the reference E11 ozonation process, we studied the contribution of different materials and electricity for various environmental impacts (Figure 5). We observed that tap water and sodium hydroxide had a negligible share in the environmental impacts. Electricity contributed greatly to the environmental impacts, such as ionizing radiations, water resource depletion, and material depletion. Liquid oxygen contributed greatly to climate change and freshwater ecotoxicity, and, to a lesser extent, ionizing radiation. Phosphoric acid contributed to the human toxicity and freshwater ecotoxicity.

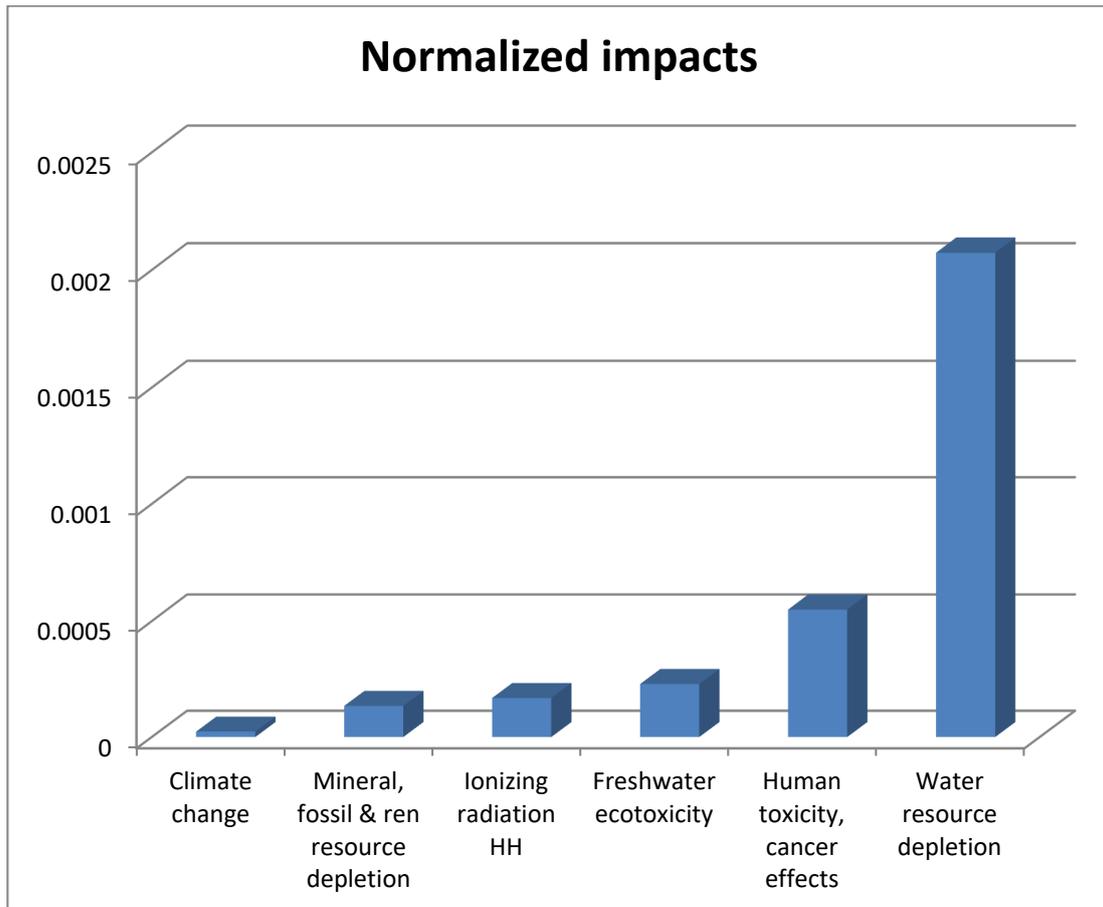


Figure 4. The life cycle assessment (LCA) impact indicators normalized for the ozone-assisted decolorization process E11.

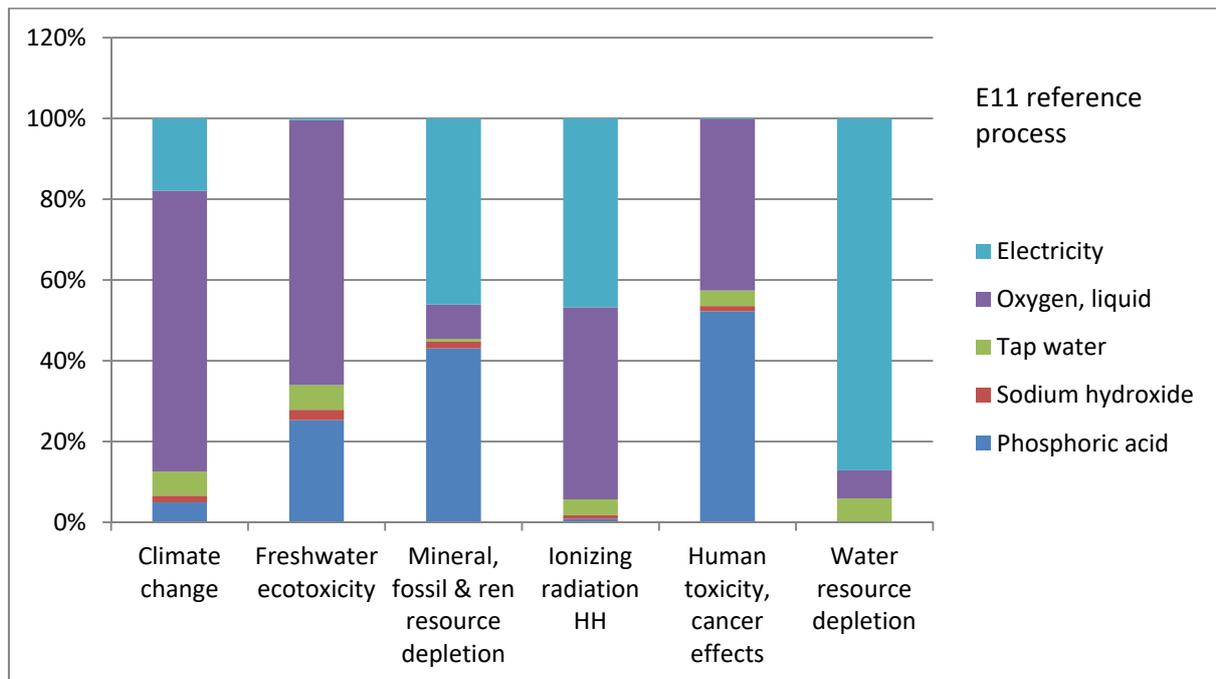


Figure 5. Contribution of different materials and electricity to various environmental impacts: reference process.

Electricity and oxygen formation are the main contributors to environmental impacts. This is related to the ozone generation. Indeed, the main electricity consumption was the ozone generator.

#### 4.2. Process Optimization Regarding Environmental Impacts

As we observed, the environmental impacts were caused by the reference process, and our aim herein was to find the best conditions in terms of the process optimization so that we could minimize such environmental impacts. The inventories for each experiment were calculated according to Section 2.3 (Table 5).

**Table 5.** Inventories for the ozone experiment.

Sr. No.	O <sub>2</sub> Required	Electricity	Phosphoric Acid	Sodium Hydroxide
	kg	kWh	g	g
E7	0.214	0.211	3.72	1.96
E8	0.214	0.211	6.86	3.6
E10	0.357	0.252	3.72	1.96
E11	0.357	0.352	6.76	3.55
E12	0.357	0.252	6.86	3.6
E13	0.214	0.151	6.76	3.55
E14	0.214	0.151	6.76	3.55
E15	0.214	0.151	6.76	3.55
E16	0.214	0.151	6.76	3.55

When treatment time decreased (Tables 1 and 5), as was the case for experiments E7 and E8, we observed that there was a reduction in the required electricity and O<sub>2</sub> input with very good color stripping.

When the ozone concentration was reduced, such as in experiments E10 and E12 (Tables 1 and 5), we observed that there was reduction in the electricity compared with the reference process. Moreover, we observed very good color stripping.

To take into account both the O<sub>3</sub> concentration decrease and the time reduction, the midpoint experiments of the statistical model (e.g., experiments E13–E16) were selected (Table 5). We clearly observed that the required electricity and O<sub>2</sub> input were less than the reference process. Color stripping was a little bit worst but decolorization still seemed significant.

#### 4.3. Introduction of the LCA Results

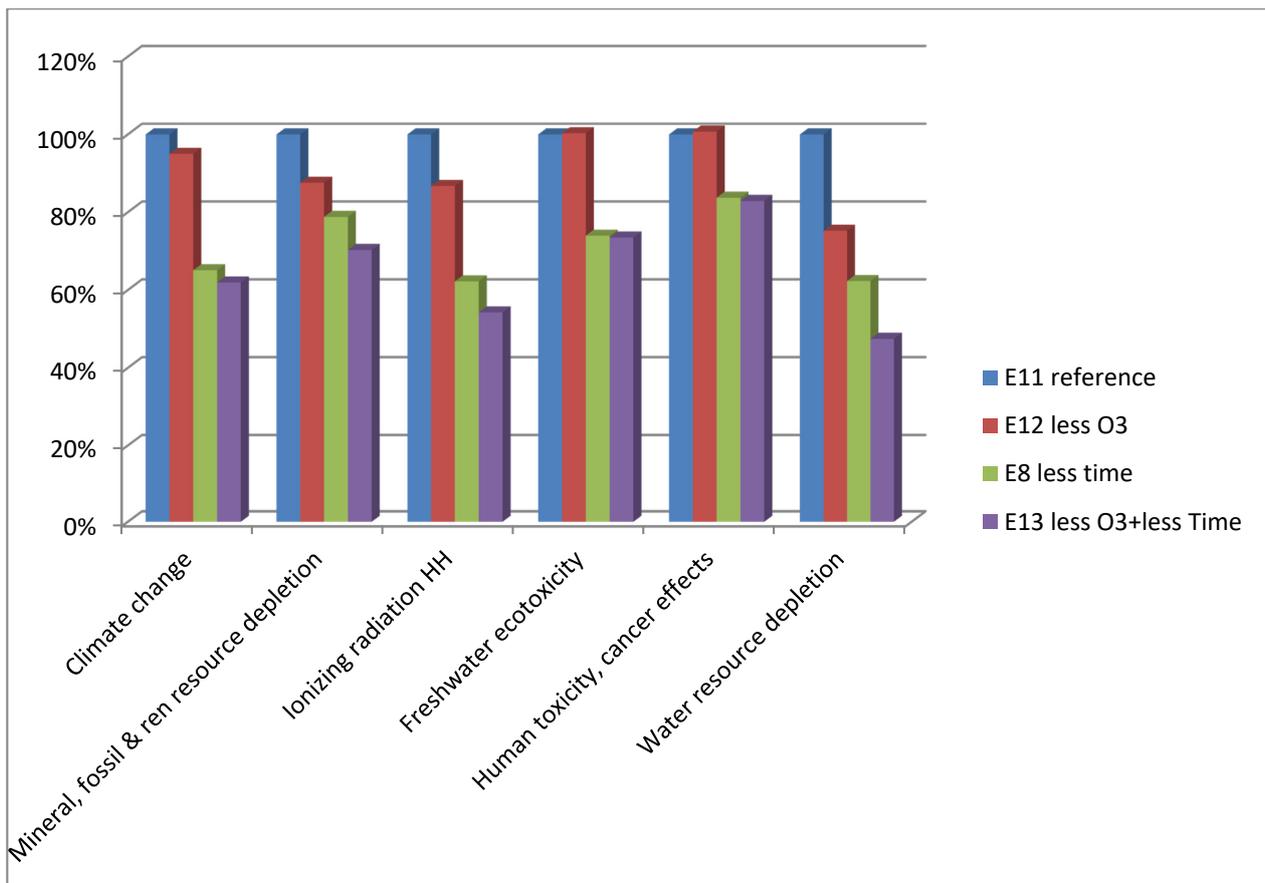
Based on the characterized results, we observed that the E13 ozonation process was preferable (Table 6). The largest differences in the impacts were observed between the reference process (E11) and the midpoint of the experiments (E13).

**Table 6.** Characterization values of the impact categories for the E11 reference, and the E12, E8, and E13 processes.

Impact Category	Unit	E11	E8	E12	E13
Climate change	kg CO <sub>2</sub> eq	0.21388189	0.13921773	0.20321215	0.13248698
Mineral, fossil, and ren resource depletion	kg Sb eq	0.00001360	0.00001072	0.00001191	0.00000956
Ionizing radiation HH	Kbq U <sub>235</sub> eq	0.19096354	0.11885205	0.16565425	0.10358892
Freshwater ecotoxicity	CTUe	2.01311521	1.48916748	2.01922845	1.47962330
Human toxicity, cancer effects	CTUh	0.00000002	0.00000002	0.00000002	0.00000002
Water resource depletion	m <sup>3</sup> water eq	0.16857161	0.10504289	0.12684691	0.08000251

Figure 6 represents the LCA results for the E11 reference, as well as E12, E8, and E13 processes. Here, the reference ozone process (E11) was compared to the different optimized processes (E8, E12, and E13). Table 6 shows that the optimized processes E8 and E13 had much lower impact values than E12 process for environmental impacts such

as climate change, ionizing radiation HH, and water depletion. The reference process E11 had the highest environmental impacts. In our LCA study based on normalized results, the atmospheric impacts, especially water resource depletion, exhibited the poorest performance among every environmental impact category. The reason could be attributed to the ozonation process setup by utilizing a large amount of water. When we observed the midpoint of the experiments, we saw that E13 had lower impacts than the reference, which used less liquid oxygen for the ozone generation and less electricity, thus reducing the overall environmental impacts. However, we obtained less color stripping, as already discussed. (Tables 1 and 6).



**Figure 6.** Comparative LCA results (normalized values) for the E11 reference, as well as the E12, E8 and E13 processes.

## 5. Discussion

So depending on the color specifications, the optimum value could be selected focusing either on the color stripping quality or on the environmental impact. If a color stripping of 94% is enough for example before dark dyeing, then the best conditions would have the lowest impact. The results obtained with the optimum conditions were good and comparable to the literature. Previous studies have shown that the reactive black 5 dyed cotton fabrics were color stripped with 96.1% and 94.4% of the stripping percent, via the electrochemical method [19].

The environmental impact of the ozone-based decolorization process was primarily caused by water use and energy consumption. The reactor utilized operates at higher material to liquor ratios; as the reactor we have used is not dedicated to textiles. The reactor design needs to be improved in order to increase the amount of fabric that could be introduced for the treatment. Since the volume of the water in the reactor was large, this also resulted in the high consumption of chemicals and auxiliaries.

This study shows that electricity is very important. In fact the overall environmental impact depends on the electricity mix and in France the electricity mix has lot of nuclear energy and that's the reason we have high ionizing radiation HH impacts. Impact categories are sensitive to the energy mix of the country. If we change the country with less electricity mix and high carbon content so we have high climate change and less ionizing radiation impact.

Moreover, the reactor utilized large amount of ozone and thus the liquid oxygen which is needed for the production of ozone. Thus, the ozone generator was also a contributor, as discussed previously. In previous studies available on wastewater treatments, the research findings showed that the ozonation process adds a 6% greater impact on climate change. This is attributed to the liquid oxygen and electricity production associated with the ozonation process [20]. In another study on the application of the LCA to the Kraft pulp industrial wastewater treatment via different advanced oxidation processes, it clearly depicted that ozonation accounted for a higher environmental impact, owing to energy consumption produced by the oxygen and ozone [21]. These results coordinate with our study. The results in this study showed that combining the ozonation with UV-A light decreased the environmental impact by about 40% [21]. In a similar study on the analysis of the advanced oxidation process, results showed that high energy consumption was a great drawback in the ozonation process [22].

## 6. Conclusions

Considering the technique utilized for the decolorization of cotton textiles using the “gate-to-gate” LCA tool, we discerned the environmental profile of the process and the hotspots associated with it.

For the ozone-assisted treatment, the electricity and oxygen formation for ozone generation were major contributors for the environmental impacts. This could be attributed to the ozone generation process, which utilized liquid oxygen and included electricity consumption due to the ozone generator.

The environmental impacts can be reduced with regards to reference process by decreasing the ozone input, decreasing the treatment time and by simultaneously decreasing the treatment time and the ozone input. However, this change in the ozonation parameters had an impact on the color stripping %.

For the ozone-assisted process, energy consumption and wastewater (pollution) related impacts were higher. “Water depletion” and “human toxicity cancer effects” were higher for the selected impact categories. We can reduce the impacts by reducing the liquor use in the ozonation process. The results obtained from the LCA study of the “gate-to-gate” provide necessary solutions that could reduce impacts, find possible solutions, and remodify the technique or process.

This study paves a route to use the ozone-based process for textile processing and allied industries at an industrial scale. It also encourages us to think and develop technologies for industrialists looking for sustainable and environmentally friendly alternatives with lower ecological impacts. Studies on the financial aspects of the process could also be an interesting research area. For future study, the LCA with different textile decolorization methods might be assessed.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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