Strategies for reuse and recycling of water and effluents in pulp and paper industries

Estratégias de reuso e reciclo de águas e efluentes em indústrias de celulose e papel

Estrategias para la reutilización y reciclaje de agua y efluentes en industrias de pasta y papel

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Abstract
Currently, concerning to the renewable resources, the decrease in the consumption of raw materials and the research for sustainable operations have been emerging as an urgent demand in all segments of the market, emphatically including, the industry. Among the most diverse industrial segments, the pulp and paper industry is responsible for the consumption of large volumes of inputs, including water. Due to the high uptake of water from water courses and the high volume of effluent discharge, some alternatives have been investigated in order to reduce the impacts caused by this activity. Alternatives are reported for the improvement of water management and wastewater minimization aiming to an integrated way to shut down the water circuits of the production plants. For this closure step, resource management solutions are presented, such as partial water reuse, use of oxidative processes, treatments by using membranes, and enzymatic processes, which reveal techniques that can be sustainably applied to industrial processes, meeting the demands of society and fulfilling the Sustainable Development Goals, therefore preserving the environment and improving life's quality of populations. In this context, this study aims to analyze, through a systematic review of previously established Brazilian databases, the alternatives and forms of reusing and recycling of water in industrial processes, focusing on closing circuits, with the characterization, benefits, and harms, of the actions performed by the pulp and paper industries.
Keywords: Water management; Paper; Cellulose; Effluent.

Resumo
Atualmente a preocupação com os recursos renováveis, a diminuição do consumo de matérias primas e a busca por operações sustentáveis tem se apresentado como uma demanda urgente em todos os segmentos do mercado, incluindo de forma enfática, a indústria. Dentre os mais diversos segmentos industriais, a indústria de celulose e papel é responsável pelo consumo de grandes volumes de insumos e matérias primas, incluindo a água. Devido à elevada captação de água dos cursos hídricos, e alto volume de descarte de efluentes, o setor vem buscando alternativas para a redução dos impactos causados por sua atividade. Em meio a estas alternativas encontram-se o reuso e reciclo de efluentes, visando de maneira integrada o fechamento dos circuitos de água das plantas produtivas. Para este fechamento, apresentam-se soluções de manejo do recurso como reuso parcial, utilização de processos oxidativos, tratamentos por membranas e processos enzimáticos que revelam técnicas simples e aplicáveis aos processos industriais de maneira sustentável, atendendo as demandas da sociedade e os Objetivos do Desenvolvimento Sustentável, de forma a preservar o meio ambiente e a melhorar a qualidade de vida das populações. Neste contexto, este estudo tem como objetivo analisar, por meio de uma revisão sistemática de bases de dados brasileiras previamente estabelecidas, as alternativas e formas de reaproveitamento e reciclagem da água em processos industriais, com foco no fechamento de circuitos, com a caracterização, benefícios e malefícios, das ações realizadas pelas indústrias de celulose e papel.
Palavras-chave: Gestão de água; Papel; Celulose; Efluente.

Resumen
Actualmente, en lo que respecta a los recursos renovables, la disminución en el consumo de materias primas y la investigación para operaciones sustentables se perfilan como una demanda urgente en todos los segmentos del mercado, incluyendo enfáticamente, la industria. Entre los segmentos industriales más diversos, la industria de la celulosa y el papel es responsable por el consumo de grandes volúmenes de insumos, incluida el agua. Debido a la alta captación de agua de los cursos de agua y el alto volumen de descarga de efluentes, se han investigado algunas alternativas para reducir los impactos que genera esta actividad. Se informan alternativas para la mejora de la gestión

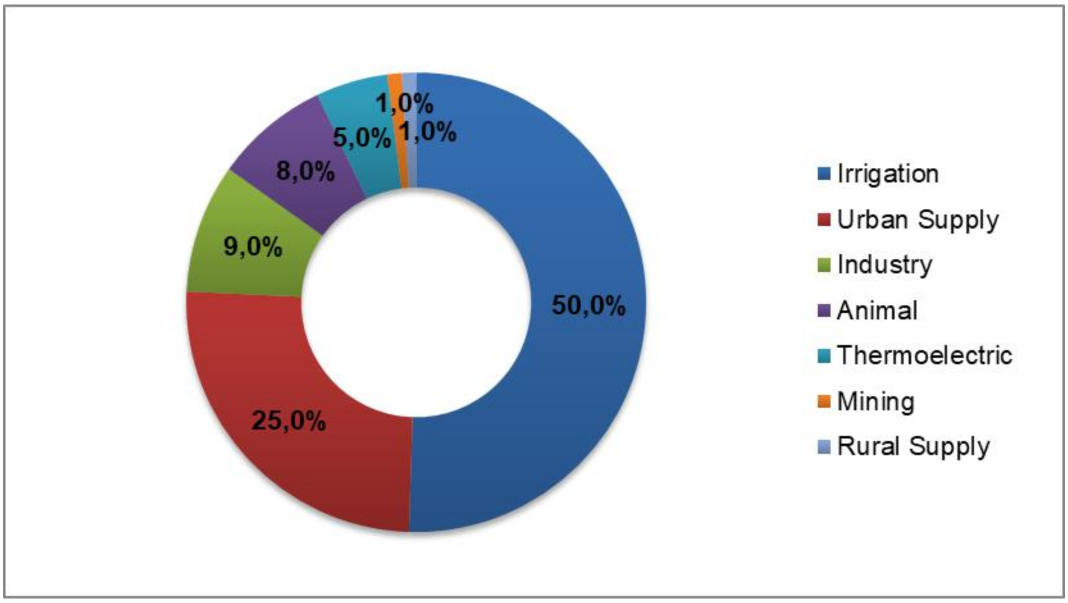
del agua y la minimización de las aguas residuales con el objetivo de cerrar de forma integrada los circuitos de agua de las plantas de producción. Para este paso de cierre, se presentan soluciones de gestión de recursos, tales como reúso parcial de agua, aprovechamiento de procesos oxidativos, tratamientos mediante membranas y procesos enzimáticos, que revelan técnicas que pueden ser aplicadas de manera sustentable a los procesos industriales, atendiendo las demandas de la sociedad y cumpliendo los Objetivos de Desarrollo Sostenible, preservando así el medio ambiente y mejorando la calidad de vida de las poblaciones. En este contexto, este estudio tiene como objetivo analizar, a través de una revisión sistemática de bases de datos brasileñas previamente establecidas alternativas y formas de reutilizar y reciclar el agua en los procesos industriales, centrándose en el cierre de circuitos, con la caracterización, beneficios y perjuicios, de las acciones de las industrias de celulosa y papel.
Palabras clave: Gestión del agua; Papel; Celulosa; Efluente.

1. Introduction

Water, a natural resource necessary for the maintenance of life and the progress of civilizations is, since the beginning, related to human development and its evolution. The strengthening of the population of different territories and the advance of agropastoral activities are influenced by the location of watercourses (Mierzwa & Hespanhol, 2005).

Just as water is necessary to meet the basic needs of populations, several other activities depend on and/or require significant amounts of this resource. According to studies carried out by the National Agency for Water and Basic Sanitation (ANA, 2021), on the Conjuncture of Water Resources in Brazil, in 2020, water consumption by activity represented an amount of 1947 m³/s, which came from Brazilian hydrographic basins. Figure 1 shows water consumption in percentage, segmented by sectors of use, in 2020.

Figure 1. Water consumption in Brazil in 2020 subdivided into sectors.



Source: adapted from the National Water Agency (ANA, 2021).

As shown in Figure 1, the three largest consumers of water are the Irrigation, Urban Supply, and Industry sectors, with a 56% growth trend in consumption over the next 20 years, which represents an increase of 61 million m³ of water captured daily from Brazilian watersheds (ANA, 2021).

With the increase in the consumption of natural resources and the rising concerns related to environmental degradation and social well-being in recent decades, the 2030 Agenda for Sustainable Development was established in 2015, an agreement signed and adopted by the 193 Member States of the United Nations (UN) (ECAM, 2022). The program is composed of 17 Sustainable Development Goals (SDGs) and 169 global action targets, covering, in an integrated manner, the environmental, economic, and social dimensions of sustainable development (Brasil, 2022). Among the 17 SDGs, Goals 6 – Clean Water and Sanitation, 12 – Responsible Consumption and Production, and 14 – Life Below Water are directly correlated with the usage of water resources by both society and the industry (Brasil, 2022).

In industry, the water used is related to the transformation of raw materials into consumer goods, which can be incorporated into the final product or act as an auxiliary in the production process (Mierzwa & Hespanhol, 2005). In the pulp and paper industry, in particular, water is a fundamental input in the production process, where part of the captured volume is integrated into the final product. In a study carried out by Bachmann (2009), the specific water consumption (m^3 of water/ton of product produced) of 28 paper mills from different segments was evaluated. According to the author, a large dispersion of values was identified, ranging from 3.4 to 91.3 m3/t of paper. The paper manufacturing sector consists of a large effluent emitter considering the volume of water captured and the low amount of incorporated water to the product (5 to 9% of the total captured volume) (Oliveira, 2012). To overcome this situation, new alternatives and technologies are being proposed to meet the imminent demands in production processes. One of these strategies consists of closing the water circuits on the industrial plants, aiming at the reuse of these effluents via total or partial decontamination and subsequent reutilization in the production of pulp and paper.

In this context, this study aims to analyze, through a systematic review of previously established Brazilian databases, the alternatives and forms of reusing and recycling of water in industrial processes, focusing on closing circuits, with the characterization, benefits, and harms, of the actions performed by the pulp and paper industries.

2. Methodology

This study addresses a systematic review, with a summary of the evidence related to the topic, through the application of explicit and systematic research methods, critical appreciation, and synthesis of selected information (Sampaio & Mancini, 2007). The systematic review, as discussed by Brizola and Fantin (2016), makes use of previously published studies as a way of understanding the available evidence on the subject of interest, making it possible to formulate a complete view of the topic addressed. To carry out this review, the methodology used by Graffunder et al. (2020) was followed, which lists seven stages of structuring a systematic review, namely: (1) Formulation of the research question; (2) Search and source selection strategy; (3) Evaluation of studies; (4) Data collection; (5) Data analysis and presentation; (6) Interpretation of the data obtained; (7) Revision update and enhancement.

Considering this structure, this research was guided by the following question: “What alternatives are available for recycling and reuse of water and effluents in Brazilian pulp and paper industries?”.

For this study, electronic research was carried out from the databases of ‘Portal de Periódicos da CAPES’, the Brazilian Digital Library of Theses and Dissertations (BDTD), and the National Institute of Industrial Property (INPI), covering the period between January 2001 and January 2022. The choice of this period can be justified by the constant technological advances in the pulp and paper area in Brazil, in addition to the growing concern with water resources, accentuated by the water scarcity and energy shortages that occurred in the country in the years 2001 and 2021.

In the systematic search strategy, conducted in February 2022, three different groups of descriptors were used, as shown in Table 1.

Table 1. Descriptors used for electronic research.

Descriptors		
Groups	**Search words**	
Group 1	Effluent	Paper
Group 2	Reuse	Paper
Group 3	Water recycling	Paper

Source: Authors (2022).

As an eligibility criterion, the descriptors that returned the largest number of published works were initially chosen. Subsequently, duplicate works were excluded and the titles of the studies were analyzed, considering their correspondence with the research objective. The abstracts of the works that showed correspondence were overviewed and then the respective complete reading was effectuated to assess whether the content was consistent with the objective proposed in the systematic review or not.

3. Results and Discussion

Through the research, using the descriptors introduced above (Table 1), the results presented in Table 2 were obtained.

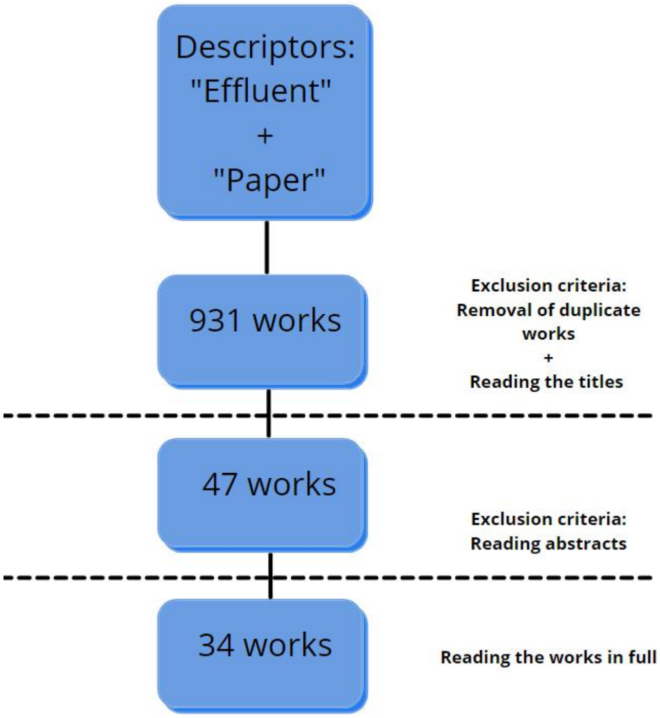
Table 2. Research results with descriptors according to the number of published works.

Descriptors		Periódicos CAPES	BDTD	INPI	TOTAL WORKS
		No. of works	No. of works	No. of works	
Effluent	Paper	612	305	14	931
Reuse	Paper	521	231	10	762
Water recycling	Paper	87	45	0	132

Source: Authors (2022).

To analyze the works, the descriptors "Effluent" and "Paper" were picked due to the return of 931 works. In this way, the exclusion of works that could potentially contribute to this review would be minimized. From the total amount of works identified in the CAPES, BDTD, and INPI Periodicals databases, duplicates were removed and the titles, abstracts, and, finally, the integrity of the remaining works were read. Figure 2 shows the works selection flow.

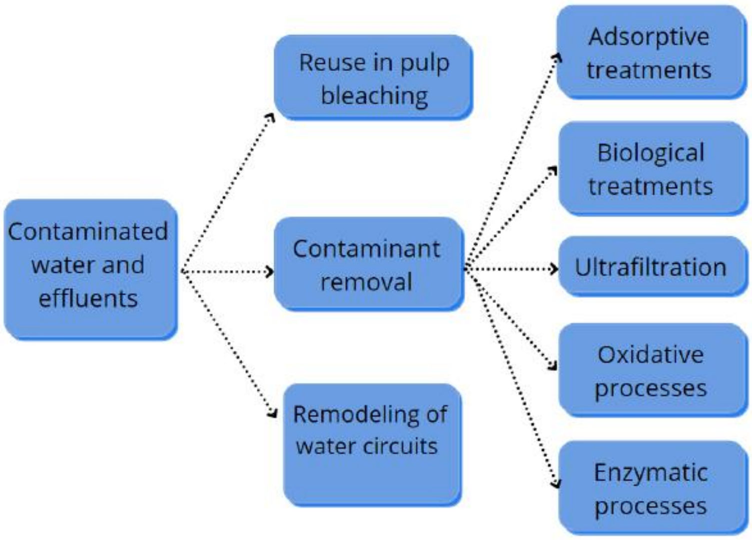
Figure 2. Selection process of works based on the descriptors "Effluent" and "Paper".



Source: Authors (2022).

Considering the works related to this systematic review, it is noticeable that the reuse of water, for the most diverse purposes, has increased in all segments of the industry, including the pulp and paper branch. In this manner, the proposed forms of water treatment for reuse are different, as this is dependent on its primary use and the contaminants added during its employment. Figure 3 shows the main forms of water and effluent reuse in the paper sector identified in this systematic review.

Figure 3. Main forms of water and effluents reuse in the paper sector.



Source: Authors (2022).

The reuse of water in the pulp bleaching process has been widely studied due to the high consumption of water, reaching values between 20 and 90 m³/t of dry pulp produced (Suhr et al. 2015). The total or partial closure of water circuits in cases of pulp bleaching is seen as an attractive option for both reducing water use and effluent generation (Santos, 2018).

As reported by Batista (2014), the practice of water recycling must be conducted cautiously to avoid incrustations in equipment and changes in the optical coordinates of the pulp, which can result in the quality variation of the product and compromise its later use. According to the author, the bleaching process requires the removal of the majority of suspended and dissolved solids, since the presence of these contaminants leads to increased consumption of reagents necessary for the appropriate whiteness of the pulp. As a result of the partial closure of the bleaching circuit, with different recycle proportions (25, 50, and 100%), a drop of 0.5 to 4.6% in the final brightness of the bleached pulp is reported.

According to Andrade (2006), when reusing the effluent from the paper machine (white water) in pulp bleaching, a reduction of approximately 13.5% in the use of freshwater was observed when considering the effect of the amount of organic matter present and higher than 30% if the presence of these contaminants is not considered.

Strategies for removing contaminants from effluents of paper industries have been addressed in countless works with the intent of avoiding disposal in water bodies; however, in a way that is not directed towards their total reuse. Studies focused on the treatment of effluents in the paper industry are presented in an applied form in some Brazilian industrial plants. Loures (2007) depicts the anaerobic treatment of white water from the paper machine using conventional bioreactors and membrane bioreactors, which provide about 20% of hardness removal. Nevertheless, an increase in pH and electrical conductivity of the effluents is observed, factors that can interfere in the stages of paper production. As stated by the author, for each production process, a certain ideal working pH is identified, and the same is observed regarding electrical conductivity; because when high, it results in increased consumption of chemical additives. In contrast to the anaerobic treatment, Sousa (2008) reports the use of thermophilic aerobic treatment, with a reduction of approximately 95% in the biochemical oxygen demand (BOD), 100% in turbidity, and total suspended solids, and 20% in effluent hardness of the paper machine. According to the writer, these data would indicate its reuse in sealing vacuum pumps, washing felts and presses, cleaning the floor, and washing shower screens, however, restrictions on its use in the rest of the process are pointed out because of the hardness parameter.

In addition to biological treatments, Bon (2017) applied a biological additive, called "Component E", in aerobic and anaerobic processes, identifying variation in the physicochemical characteristics of the studied effluent, and interference in the efficiency of the removal of organic load due to the presence of contaminants. As the author affirms, the anaerobic treatment showed practically zero organic matter removal, while the aerobic treatment presented results of BOD removal of 88% in the RA3 configuration (aerobic reactor inoculated with 300 mL of "Component E" and 900 g of cattle manure), being, therefore, the best working condition found. It was shown that this inoculum could be used for effluents' treatment in the pulp and paper industries.

Another method intended to remove phenolic and lignin compounds from effluents of paper industries, proposed by Araújo (2013), consists of using different tertiary treatments for effluents using MBBR reactors (Moving Bed Biofilm Reactors) followed by biological treatment in aerated ponds. Better performance in terms of COD removal and better cost-effectiveness were identified in physicochemical treatments given the fact that a high load of contaminants presents influences the response in the case of oxidative processes, due to the presence of suspended materials that inhibit the action of chlorine dioxide. Vanzetto (2012), also making use of MBBR reactors, reports on the influence of changing the organic loading rates (OLRs) on its removal from the effluent. Under the operational conditions defined, rates of 47.9% and 94.2% of COD and BOD5 (biological oxygen demand) removal, respectively, were identified, being considered a promising result for the treatment of effluents.

In the patent registered by Ristolainen et al. (2017) is presented an alternative treatment for calcium, an element that is difficult to remove and which becomes an aggravating factor for the reuse of effluents in paper machines. For the removal of this contaminant, the authors proposed the insertion of exhaust gas (CO_2) in the primary effluent of a pulp and paper mill, with the consequent formation of calcium carbonate. The calcium carbonate can then be separated from the effluent and reduced to calcium oxide (CaO) in a lime kiln, enabling its reuse in the pulping process and making the effluent suitable for recycling in the form of process water.

In addition to the alternatives already mentioned, adsorptive treatments are reported for water reuse, such as the use of clay and activated carbon to remove color and organic matter, a method applied by Hinojosa (2014). In the treatment via adsorption reported by the author, the results demonstrate the potential of activated carbon as an adsorbent material, responsible by the removal of color and organic matter, being used in the tertiary treatment of biologically treated Kraft process effluent. Bender, et al., (2019) proposed the evaluation of the combined processes of coagulation, flocculation, and sedimentation, in addition to adsorption on granular activated carbon for the removal of color and phenol from the effluent of the pulp and paper industry. The complete cycle treatment, respecting all the previously mentioned steps, demonstrated contaminant removal rates above 90%, except for COD data, of about 64%.

Other authors report the use of flocculation and coagulation treatments with aluminum sulfate and ferric chloride, associated with photodegradation, electroflotation, electrodialysis, and oxidative processes. In the photodegradation processes reported by Barros and Nozaki (2002), Marques (2014), and Rodrigues (2007), it was identified that a previous flocculation/coagulation steps are extremely important to reduce the irradiation time of the photocatalytic treatment. In addition, depending on the catalyst used, this degradation technique provides about 60% of COD reduction and of effluent toxicity. The electrodialysis, proposed in the form of a patent document by Vogel and Reyes (2021), is used for the separation of inorganic salts from effluent previously exposed to conventional primary, secondary and tertiary treatments, allowing its reuse in the pulp and paper mill. As claimed by the authors, advantages to the industry are pointed out, such as the recovery of water for use in periods of drought, the reduction of effluent discharges into bodies of water, the recovery of useful chemical products for the treatment of effluents or factories of pulp and paper and, finally, the reduction of water consumption per ton of product produced. On the other hand, considering the oxidative processes to water reuse, Souza et al. (2017) highlight that the use of the Fenton process resulted in a reduction of 74.9% in COD and 87.2% in total phenols. According to Tambosi (2005), in combined coagulation and flocculation processes followed by UV radiation and application of hydrogen peroxide (H_2O_2), 60% of total COD removal was obtained. Giroletti (2017) using UV/H_2O_2 technology, attained COD reduction of 84% with the application of 100 to 200 mg.L^{-1} of H_2O_2, and 91 % of BOD_5 reduction with the application of 150 and 200 mg .L^{-1} of H_2O_2. Vaz (2004) used the same treatment technology, highlights COD and total phenols removal of 99 and 95%, respectively, and about 80-95% of lignin degradation.

Ozone was also applied as an advanced oxidative process by Medeiros (2008), being obtained a color reduction of 98.7% for primary effluent, and values between 44.4 and 77.8% for different effluents from the alkaline extraction sector. The author also pointed out the increase increase of biodegradability (ratio between BOD_5/COD) from 0.18 to 0.35 in primary effluent and from 0.07 to 0.15 for alkaline extraction effluent, showing that oxidative treatment consists in an alternative to minimize the contamination of effluents.

Ferreira (2006), using the electroflocculation technique with aluminum and iron electrodes, showed that the treatment with aluminum was more effective, being attained average values of 93, 99 96 and 26 % of reduction for turbidity, BOD, COD, and electrical conductivity of the effluent, respectively.

The application of ultrafiltration membranes in paper industry is a fact since one of the main sources of effluent in this sector is white water from paper machines. Oliveira (2003), in the use of ultrafiltration for white water recycling, emphasizes the obtention of 100% of suspended solids removal and 84% of reduction in effluent turbidity, without removing the calcium, which was later treated by precipitation. Aiming to calcium removal, Sousa et al. (2011), report values between 75 and 87% of calcium recovery from the effluent by precipitation with sodium oxalate. However, according to the authors, this effluent ought not to be used in heat exchanger equipment, due to residual concentrations of oxalate, calcium, and organic matter, which can result in the formation of incrustations. Furthermore, no changes were observed in the post-precipitation conductivity values of the effluent, which is a positive point for the subsequent application of this reused water. Coutinho and Woodhead (2018) highlight the use of ultrafiltration associated with calcium hydroxide chambers, and subsequent clarification, in the obtention of an effluent free of COD and chromophore compounds which would provide a color in the effluent from the pulp and paper industry.

Another technology presented was "Lo-hardness Ultrafiltration – LhU" - Softening of White Water Through Chemical Precipitation and Conjugate Ultrafiltration, filed as a patent by Milanez et al. (2013). This system is characterized by the complete removal of hardness and total suspended solids from industrial effluents using chemical precipitation with the elevation of the effluent's pH and subsequent ultrafiltration, utilizing submerged polymeric membranes with an average pore size of 0.2 μm. In this way, all material of larger dimensions is retained in the membranes, achieving simultaneous removal of 99% of hardness and 100% of turbidity and suspended solids in the industrial white water.

It has been observed in scientific publications as well as in the industry, forms of restructuring and/or evaluation of the water circuit, searching alternatives to the better use of the available resource. Moreira (2010) evaluated the reduction of kraft pulp washing water by analyzing the water circuit. The author identified an increase in non-processable elements, such as K, Mg, Cl, Si and Ca, with the increase in the rate of circuit's closure. This closing of the circuit provided savings of 11.4% in the volume of freshwater injected into the process, without implications in the quality of the product. Similarly, Nunes (2007) presented a study for the the minimization of water consumption in a paperboard machine using hydric diagnostic. The algorithm developed to the determination of the reuse potential of effluents adopted different levels of priority, such as direct reuse, direct reuse with dilution, recycling, and post-treatment reuse. As a result, 66% of reduction in freshwater consumption and 54% of reduction in the total effluent generated were identified. In turn, Dolny (2011) reports a study to minimize white water effluents from a paper machine using hydric diagnostic and subsequent analysis for the characterization of white water. The author verified a linear relationship of dependence between the flow rate of white water effluent and the loss of paper fibres in the process. In this sense, the strategies used to the reduction of the white water effluent do not change the properties and characteristics of the final product. In addition, it was identified 14.8% of reduction in the consumption of treated water fed to the paper machine (about 70 m^3/h) and a 13.7% of reduction in fiber loss, which may represent savings in fibers of approximately US$ 108.000.00/year for the studied industry.

When it comes to technological evolution in the treatment of water and effluents with a biotechnological bias, research related to the use of fungal enzymes has been gaining ground in different industrial segments. Kumar et al. (2015) presented a process for the aerobic treatment of pulp mill wastewater aiming to the color reduction with the use of a bacterial strain. In the study, the ability of the isolated bacteria for the color reduction of the effluent in a reproducible manner was observed. Moreover, the culture in question is non-pathogenic and can be grown by simple nutritional means. Therefore, it can be presented as an alternative system to the treatment of effluents and recycled waters of complex compositions such as those generated in the pulp and paper industry.

In the patent filed by Dillon et al. (2018), the use of *Pleurotus Sajor-caju* fungus is described in cultivation at increasing concentrations of phenolic compounds present in industrial effluent. After the fungus cultivation, there is a step of aerobic treatment of the effluent by the action of activated sludge. The results obtained by the authors demonstrate that the effluent from the pulp and paper industry, when used to favor the fungal growing, presents reduced toxicity to bacteria from activated sludge, thus enabling the removal of organic matter. In addition to the work by Dillon et al. (2018), Bonfanti (2014) reported the use of the same *Pleurotus* strain for the treatment of effluents from the pulp and paper industries. In this work, it was observed a reduction in total phenols content and the stability of the chromophore compounds that give the effluent its color, which are not removed by the enzymes action.

In addition to the genus *Pleurotus*, Vilela (2006) studied the *Aspergillus* sp cultivation for the laccases production which can be applied in the treatment of effluents from the paper industry. As a result, it was demonstrated the action of immobilized enzymes in chitosan in the biotransformation of the compounds of interest. Removal percentage of about 65% of low molecular weight phenolic compounds, 65% of the total phenolic compounds, and 60% of the effluent's color under study were attained. Machado (2017) reports the use of commercial laccases in the evaluation of the treatability of kraft cellulose effluent by biological process. According to the author, it was found that the best-operating conditions resulted from the application of the aerated biological system, associated with the use of laccase at 3.9 U/mL of activity, pH 4.0, and a temperature of 37 °C. Under these favorable conditions for catalysis, 52% of COD removal, 20% of color and 30% of lignin compounds reduction were observed.

4. Final Considerations

This review shows that the reuse of water and effluents in the paper and cellulose industries has gradually been applied in productive processes due to environmental concerns, low water availability and large volumes of consumption in this industrial segment. There are countless treatments options for effluents and water from process, and in this same proportion, several are the forms of using them after the execution of these treatments. With the advance in scientific development and, in this direction, biotechnology, novel processes have been implemented and applied in the treatment of effluents. The use of different types of microorganisms and/or enzymes for removal of different compounds of hard degradation, such as lignin and other contaminants, can result in acceptable levels of reuse of process water in all stages of production, eliminating the necessity of sectored treatment procedures, as presented in the works covered in this systematic review. As a prospect for future research and work, it is suggested to deepen the reuse alternatives in the paper manufacturing process, since the main responsible for the reuse and recycling of water and effluents in this segment are the pulp production plants. In addition, it is also suggested that biotechnological research be carried out to increase the quality of water and effluents, in order to use enzymes produced by microorganisms in these treatments.

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