



# Characteristics and treatment of Brazilian pulp and paper mill effluents: a review

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Received: 30 January 2022 / Accepted: 25 July 2022 / Published online: 6 August 2022  
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**Abstract** Pulp and paper industries are very important for developing the Brazilian economy. During production processes, many effluents are generated with high polluting potential. The objective of this study is to conduct an extensive literature review on the characteristics of effluents and treatment forms adopted by Brazilian mills in this industrial sector. Most consulted studies address raw (without treatment) and secondary (after biological treatment) effluents, considering their main characteristics like pH, chemical and biochemical oxygen demands (COD and BOD, respectively), color, solids, organochlorines, toxicity, estrogenic activity, and phenols. Raw effluents differ considerably in composition, depending on the type of paper produced, the pulping process employed, and other steps, like pulp bleaching. Raw effluent characteristics indicate that this effluent cannot be directly disposed of into water bodies, because it does not comply with federal and state disposal standards. Secondary effluents normally

comply with Brazilian legislations, although some studies have reported COD and total phenol concentrations higher than disposal standards, suggesting that additional treatments are necessary. Treated effluent reuse was verified in some Brazilian mills, while its disposal in eucalyptus plantations has been considered a promising alternative for irrigation purposes.

**Keywords** Pulp and paper effluent · Environmental legislation · Organic pollutants · Toxicity · Estrogenic activity · Industrial effluent treatment

## Introduction

The pulp and paper industries are a very important part of the Brazilian economy. Most companies in this sector are located in the southern and southeastern regions of the country. In 2020, Brazil was the second largest producer of cellulosic pulp, at 21 million tons, behind only the USA, and the tenth largest paper producer, at 10.2 million tons. Even with the COVID-19 pandemic, pulp production increased at 6.1%, while paper production fell 2.8%. All raw materials for producing pulp and paper comprise eucalyptus and pine from planted forests (IBÁ, 2021).

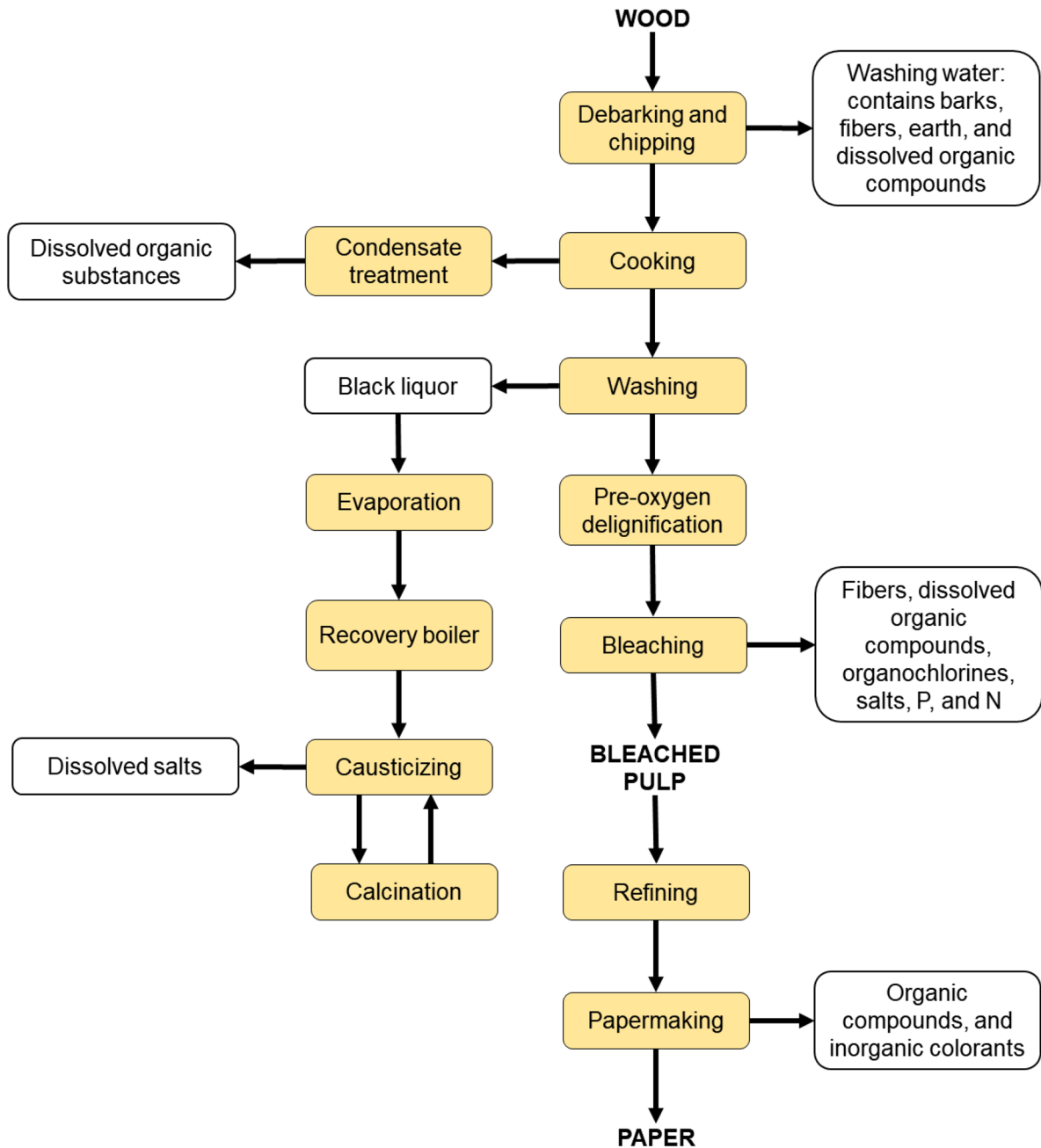
The conversion of wood fibers into cellulosic pulp and paper occurs through chemical processes, like the kraft and sulfite processes, and mechanical processes, like thermomechanical pulping (TMP) and chemithermomechanical pulping (CTMP) (Bajpai,

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2016). The most commonly used process in Brazil for producing cellulosic pulp is the kraft process (Follmann, 2017; Rocha, 2017). After debarking and chipping the wood, a pulping/cooking step at high temperature and under high pressure is carried out in

an alkaline solution containing NaOH and Na<sub>2</sub>S, followed by a separation and washing step of the brown pulp, pre-oxygen delignification, bleaching, and refining, finally resulting in the manufactured paper, as shown in Fig. 1 (Bajpai, 2016; Cabrera, 2017; Kamali



**Fig. 1** Kraft manufacturing process for obtaining bleached cellulosic pulp and paper, and effluent composition at each stage (in the white boxes). Adapted from Cabrera (2017) and Simão et al. (2018)

& Khodaparast, 2015). It is important to mention that not all mills use the last three stages.

Pulp and paper industries use many natural resources, especially vegetable fibers and water, and consume a lot of energy, especially during the cooking stage (Ashrafi et al., 2015; Bajpai, 2016; Kamali & Khodaparast, 2015). Huge freshwater consumption occurs in the process, resulting in a huge amount of effluent. Approximately 60 m<sup>3</sup> of water is demanded per ton of paper produced (Lindholm-Lehto et al., 2015). Effluents arise from virtually every step, from debarking up to paper production (Fig. 1). The bleaching stage generates the most effluents. Black liquor is not an effluent of the process, since it is treated separately for recovering chemical inputs and generating steam and energy (Ashrafi et al., 2015; Cabrera, 2017; Kamali & Khodaparast, 2015).

Effluents from the cellulosic pulp and paper industry contain organic compounds from degraded carbohydrates, lignin, and extractives, in addition to organochlorines from the bleaching steps that use chlorine-containing inputs, especially ClO<sub>2</sub> (Hubbe et al., 2016). Water quality where waste is dumped is affected by the improper disposal of these effluents if not properly treated (Furley et al., 2018). In addition, pollutants can accumulate in the sediments (Yadav & Chandra, 2018). Improper disposal leads to decreased dissolved oxygen levels and other changes in the physicochemical characteristics in water bodies, in addition to toxic effects on local biota (Ojunga et al., 2010; Tripathy et al., 2022). Most mills use primary and secondary treatment methods for effluents. Primary treatments generally use decanters to separate suspended solids via sedimentation, especially fibers. Secondary treatments are usually aerobic biological processes via an activated sludge system or aerated ponds to remove dissolved and colloidal biodegradable organic matter (Ashrafi et al., 2015; Cabrera, 2017). When necessary, tertiary treatment processes can be used, which polish secondary effluents to comply with environmental regulations and minimize impacts, although they are rarely used. When used, tertiary treatments remove COD, color, and suspended solids (Ashrafi et al., 2015; Cabrera, 2017).

There are some recent reviews in the literature that addressed solid wastes generated in the pulp and paper mill (Simão et al., 2018), while others have focused on diverse processes for treating wastewater (Ashrafi et al., 2015; Hubbe et al., 2016; Kamali

& Khodaparast, 2015; Mandeep et al., 2019), types of biological process (Hossain & Ismail, 2015), advanced oxidation processes (Hermosilla et al., 2015), and treatment through microbial fuel cells (Toczyłowska-Mamińska, 2017). As a novelty, this literature review presents a critical analysis on raw and secondary effluent characteristics from Brazilian pulp and paper companies. These characteristics will be analyzed considering the conventional treatment techniques and current legislation for discharging effluents into water bodies. An extensive research was conducted from scientific articles, dissertations, papers presented at conferences, and articles published in journals through research sites like SciELO, Google Scholar, Scopus, and the Web of Science. The key themes were “treatment of pulp and paper industry effluents in Brazil” for recent publications between 2010 and 2020. With the collected data, the values and magnitudes of commonly monitored parameters for raw and secondary effluents were analyzed. Reports on reusing treated conventionally effluents were also addressed. Furthermore, correlations between some characteristics were verified to establish predictive models to streamline decision-making for treating these effluents.

### Characteristics of raw effluent

The characteristics of raw industrial effluents are presented in Table 1, arranged according to the type of pulping process. Average values or values referring to separate samples have been presented depending on how they were reported by the consulted sources. Only mean pH data were omitted since these are presented on a logarithmic scale. When sources reported maximum and minimum pH values or single measurement values, these were considered here. Large variations among the different reported characteristics were observed, perhaps in the function of the processing steps involved, the amount of water used, or the wood and inputs used. Industries usually produce different pulps and/or papers, meaning that effluents vary significantly in terms of composition, even within the same industry. Some studies collected and characterized more than one effluent sample and from different days in the same mill. Considerable differences were observed for some characteristics, mainly BOD<sub>5</sub>

**Table 1** Raw effluent characteristics from Brazilian pulp and paper industries

Process	pH	BOD <sub>5</sub> (mg L <sup>-1</sup> )	COD (mg L <sup>-1</sup> )	BOD <sub>5</sub> /COD	Color (mg PtCo L <sup>-1</sup> )	Turbidity (NTU)	Lignin	TP (mg L <sup>-1</sup> )	TS (mg L <sup>-1</sup> )	TSS (mg L <sup>-1</sup> )	References
Kraft with bleaching	–	–	2371	–	1048	–	0.47 (280 nm)	–	–	–	Chaparro et al. (2010a, b)*
	–	386	859	0.33	668	–	0.31 (280 nm)	–	–	–	Chaparro and Pires (2011)*
	–	1039	2193	0.47	943	–	0.46 (280 nm)	–	–	–	
Kraft with bleaching	3–7	590	1583	0.37	545 (TC)/591 (AC)	–	–	0.1	5237	324	Amaral et al. (2013)*
	10.3–11.2	741	1455	0.51	357 (TC)/756 (AC)	–	–	0.07	5969	1027	
Kraft with bleaching	–	335	1145	0.29	–	–	20 mg L <sup>-1</sup>	–	–	–	Ruas et al. (2012)
Kraft with bleaching	–	1090	2627	0.41	1350	–	–	–	–	–	Cabrera-Padilla and Pires (2014)
Kraft with bleaching	7.4	694	1235	0.56	253	–	–	–	–	–	Rabelo et al. (2014)
	7.2	576	1204	0.48	238	–	–	–	–	–	
Kraft with bleaching	–	525	1217	0.43	–	–	–	–	3349	180	Castro et al. (2018a)
Kraft with bleaching	–	–	1340	–	207	–	–	–	–	–	Matias et al. (2018)
Kraft with bleaching	–	530	1309	0.40	–	–	–	–	–	185	Mata et al. (2020)
Kraft without bleaching	–	251	768	0.33	0.57 (440 nm) (TC)	–	3.53 (280 nm)	–	–	–	Vanzetto (2012)
Kraft without bleaching	–	345	1045	0.33	278	4.2	1.087 (280 nm)	–	2040	418	Giroletti (2017)
Kraft without bleaching	–	215	656	0.34	0.52 (440 nm)	51.9	4.26 (280 nm)	–	–	–	Machado (2017)
Kraft without bleaching	–	171	602	0.28	0.46 (440 nm)	–	4.50 (280 nm)	–	–	–	Castro et al. (2019)
	–	239	688	0.35	0.43 (440 nm)	–	5.14 (280 nm)	–	–	–	
Kraft	6.8	1187	2682	0.44	–	–	–	1.1	–	388	Magnanti et al. (2018)

**Table 1** (continued)

Process	pH	BOD <sub>5</sub> (mg L <sup>-1</sup> )	COD (mg L <sup>-1</sup> )	BOD <sub>5</sub> /COD	Color (mg PtCo L <sup>-1</sup> )	Turbidity (NTU)	Lignin	TP (mg L <sup>-1</sup> )	TS (mg L <sup>-1</sup> )	TSS (mg L <sup>-1</sup> )	References
Kraft	–	414	1047	0.40	0.41 (440 nm) (TC)/0.73 (440 nm) (AC)	129 FTU	0.5 (280 nm)	84	1618	–	Micheletto et al. (2019)
Without bleaching	8.1	750	1748	0.43	0.46 (440 nm, TC)/0.61 (440 nm, AC)	73.4 FTU	0.5 (280 nm)	76	954	–	
Without bleaching	10.5	338	1769	0.19	300	43	–	–	–	50	Pacheco and Soares (2011) Farias and Ferreira (2014)
CTMP	–	7674	8567	0.90	6.95 (440 nm, TC)	855	49 (280 nm)	–	–	1110	Grözner et al. (2013)
CTMP	–	6351	9992	0.65	7.8 (440 nm, TC)	1439	58.9 (280 nm)	–	–	1570	Grözner et al. (2018)
TMP with bleaching	–	2872	7266	0.40	7780 (TC)	152	–	–	5779	1771	Caldeira (2019)*
Tissue paper production	–	163	446	0.37	–	–	–	–	–	593	Brandão (2014)
Paper production	–	488	780	0.62	–	–	–	–	–	566	Morais et al. (2016)
Paper production	6.8	–	363	–	46 (TC)/475 (AC)	–	–	–	590	298	Bender et al. (2016)
Paper production	7.2	–	810	–	948 (AC)	–	–	–	–	27	Cabral (2016)
Paper production	7.1	–	753	–	224 (TC)/880 (AC)	88.9	–	–	1656	54	Prestes et al. (2016)
Paper production	7.8	–	446	–	1104 (AC)	812	–	–	1084	545	Managó et al. (2018)
Paper production	7.8	–	570	–	1760 (AC)	544	–	–	1050	410	

**Table 1** (continued)

Process	pH	BOD <sub>5</sub> (mg L <sup>-1</sup> )	COD (mg L <sup>-1</sup> )	BOD <sub>5</sub> /COD	Color (mg PtCo L <sup>-1</sup> )	Turbidity (NTU)	Lignin	TP (mg L <sup>-1</sup> )	TS (mg L <sup>-1</sup> )	TSS (mg L <sup>-1</sup> )	References
White water	6.7	–	295	–	100	9.6	–	–	700	20	Santos et al. (2020)
Not informed	9.8	–	2588	–	3124 (AC)	–	–	–	–	–	Souza et al. (2018)
Not informed	10.5	339	780	0.43	–	120	–	–	3500	–	Tilha et al. (2019)

PtCo platinum–cobalt unit, TC true color, AC apparent color, NTU nephelometric turbidity unit, FTU formazin turbidity unit, TP total phenols

\*Bleaching stage effluents

(Amaral et al., 2013; Castro et al., 2019; Chaparro & Pires, 2011; Micheletto et al., 2019).

After verifying the pulping processes, it was found that most effluents come from the kraft process, with or without pulp bleaching stage. Some sources did not mention this step as existing at the plants where effluent was collected. Few studies reported effluents from the CTMP process. The latter uses inputs like bisulfite during the pressurized steam stage and then defibrates wood chips under pressure for a certain time (Bajpai, 2016). Only one source studied TMP effluent (Caldeira, 2019). In this process, wood chips are exposed to pressurized steam for a short period of time with no chemicals, followed by refining (Kamali & Khodaparast, 2015). By contrast, some studies did not mention pulping type, but rather their characteristics, which were very similar to kraft pulping effluents. Some reported effluents came from companies that only make paper, i.e., companies that purchased processed pulp from other companies or used scraps (recycled paper). Paper machine effluent is commonly called white water (Kamali & Khodaparast, 2015; Santos et al., 2020). It is worth noting that some effluents came exclusively from bleaching steps, with or without using chlorine-containing inputs.

When evaluating raw effluent characteristics, the reported pH values were generally above 7. This is consistent with soda use in various chemical and semi-chemical pulping processes (Cabrera, 2017). Widely ranged values were observed for COD and BOD<sub>5</sub>, the most common organic matter concentration parameters, ranging from hundreds to thousands of mg L<sup>-1</sup>. There were samples with higher COD and BOD<sub>5</sub> values compared to others, above 7000 mg L<sup>-1</sup> and 9000 mg L<sup>-1</sup>, respectively. These values were observed for samples from mechanical processes. Melchioris and Xavier (2018) state that CTMP processes use less water and consequently generate less effluent, but which is more concentrated than kraft process effluent. This shows that pulping type significantly interferes with wastewater characteristics. The National Environmental Council (CONAMA) establishes that effluent treatment must remove 60% BOD<sub>5</sub>, while COD is not mentioned (Brazil, 2011). On the other hand, state environmental agencies require maximum concentrations of BOD<sub>5</sub> and COD from 40 to 180 mg L<sup>-1</sup> and from 60 to 400 mg L<sup>-1</sup>, respectively, or minimum removal levels from 30 to 95% and from 60 to 90%, respectively, depending on

the state (Morais & Santos, 2019). It appears that, in general, organic matter concentration present in raw effluent is well above established limits and requires treatment.

Other distinctions can be made beyond a brief comparison between kraft and mechanical processes. The main observed characteristics for paper manufacturing effluents are pH close to 7 and COD similar to domestic sewage in Brazil, ranging from 360 to 810 mg L<sup>-1</sup> (Budeiz & Aguiar, 2020; Silva & Mendonça, 2003; von Sperling, 2014). For effluents from the kraft process followed by the bleaching step, COD values are higher, between 860 and 2600 mg L<sup>-1</sup>, and BOD<sub>5</sub> varied between 330 and 1090 mg L<sup>-1</sup>. The pH values are slightly above 7, except for some effluents obtained from bleaching stages only, which ranged from 3 to 11. On the other hand, unbleached kraft pulp effluents had COD and BOD<sub>5</sub> values lower than 1050 mg L<sup>-1</sup> and 360 mg L<sup>-1</sup>, respectively, suggesting that the bleaching stage increases organic matter concentration in effluents.

Based on the data of organic matter concentration, the BOD<sub>5</sub>/COD ratio was calculated. This ratio indicates whether effluent is biodegradable, allowing researchers to gain approximate knowledge on the decomposition capacity of organic matter during biological treatments. A better treatment method for effluent can, thereby, be proposed (Dezotti, 2008; von Sperling, 2014). It was observed that 65% of the samples had a BOD<sub>5</sub>/COD ratio equal to or above 0.4. Ratios above this indicate that effluents have a biodegradable fraction above 40%, and therefore, biological processes are recommended in the treatment steps. Lower values mean that effluent can be treated better through physicochemical processes. Biological processes are always indicated due to two main aspects: efficiency and cost-effectiveness (Dezotti, 2008; von Sperling, 2014).

Another widely analyzed parameter is color, since these effluents are usually dark brown. Color is attributed to extractives, polymerized tannins, lignin, and its dissolved derivatives, which are released in pulping, bleaching, and recovery steps (Ashrafi et al., 2015; Hubbe et al., 2016; Lindholm-Lehto et al., 2015). Color is usually reported as being true color (TC), apparent color (AC), or not specified. True color is the color of a filtered or centrifuged sample that removes suspended colloidal materials. Apparent color is based on the raw

sample (von Sperling, 2014). It was observed that some data for color were reported simply in terms of absorbance. This hinders comparisons between samples. Turbidity is the degree to which light is obstructed due to suspended solids. There are no disposal limits for color and turbidity, according to Brazilian environmental agencies, but they are often monitored to determine the efficiency of conventional or alternative treatments.

It is common for pulp and paper effluents to quantify total solids (TS) and total suspended solids (TSS). Although maximum TS concentration is not required for disposal, some state environmental agencies limit maximum concentrations for TSS in effluents from 40 to 180 mg L<sup>-1</sup> (Morais & Santos, 2019). It was verified that most raw effluent samples were above this range. Sedimentable suspended solids (SSS) were quantified in a few studies (Amaral et al., 2013; Cabral, 2016; Farias & Ferreira, 2014; Magnanti et al., 2018), and the values varied from 24 to 97 mL L<sup>-1</sup>, well above the limit value of 1 mL L<sup>-1</sup> (Brazil, 2011). When calculating the TSS/TS ratio, it was found that TSS corresponded to less than half of the first. It appears that most solids in these effluents are dissolved and not suspended. Suspended solids comprise fibers that were lost in the bleaching stage (Hossain & Ismail, 2015). Primary treatment removes them via sedimentation. Furthermore, recovered fibers can be reused in paper manufacturing (Kossar et al., 2013; Tilha et al., 2019). Parts of the inorganic dissolved substances are sodium salts and bleaching agents (Mandeep et al., 2019), while dissolved organic substances are mostly lignin and hemicellulose fragments, and extractives, many of which are chlorinated due to the bleaching step with chlorine dioxide (Cabrera, 2017; Hubbe et al., 2016).

Lignin fragments are usually partly responsible for the COD, color, and toxicity of these effluents (Hubbe et al., 2016; Kamali & Khodaparast, 2015). The content of these substances is usually reported in terms of absorbance at 280 nm. Phenols are among the most common and toxic pollutants in industrial effluents (Wasi et al., 2013). In some studies, phenolic compounds were quantified, which is a way of reporting lignin degradation products. Total phenols (TP) are limited to 0.5 mg L<sup>-1</sup> for discharging into watercourses (Brazil, 2011). Few raw effluents had values well above this limit, although one of them exceeded 80 mg L<sup>-1</sup>.



Other physicochemical parameters were rarely quantified for effluents. Total organic carbon (TOC), for example, is another indirect parameter used for quantifying organic matter concentration (von Sperling, 2014), yet it was only reported in five studies. Values ranged from 125 to 8203 mg L<sup>-1</sup> (Giroletti, 2017; Grötzner et al., 2018; Hinojosa & Xavier, 2016; Matias et al., 2018; Ruas et al., 2012). Dissolved organic carbon was rarely quantified in studies from the same research group (Chaparro & Pires, 2011; Chaparro et al., 2010a, b). Values ranged from 335 to 953 mg L<sup>-1</sup>.

Other important parameters evaluated were adsorbable organic halogens (AOX) (Chaparro & Pires, 2011; Chaparro et al., 2010a, b; Ruas et al., 2012) and chloride concentration (Caldeira, 2019; Chaparro & Pires, 2011; Giroletti, 2017; Matias et al., 2018; Rabelo et al., 2014; Ruas et al., 2012). These pollutants are generated in the bleaching stages (Hubbe et al., 2016). It is known that the amount of AOX has been decreasing due to the replacement of elemental chlorine by elemental chlorine-free or totally chlorine-free bleaching sequences (Lindholm-Lehto et al., 2015). Chloride concentration ranged from 21 to 696 mg L<sup>-1</sup>, and AOX ranged from 16 to 23.8 mg Cl<sup>-</sup> L<sup>-1</sup>. Some organochlorines are toxic and carcinogenic, in addition to not being easily degraded by biological treatments. Such compounds can also cause endocrine dysfunction and hepatic problems in aquatic organisms (Ashrafi et al., 2015; Hubbe et al., 2016).

Phosphorus and nitrogen concentrations are not commonly quantified, since concentrations of these parameters are known to be very low in these effluents. N and P are usually solubilized in the black liquor generated in the kraft process (Ashrafi et al., 2015). It is common, however, to add N and P as a supplement for biologically treating these effluents in bench scale (Gao et al., 2016; Hooda et al., 2015;

Morais et al., 2016). Effluent treatment plants from this sector use predominantly aerobic biological processes. An ideal proportion between organic matter and nutrients for aerobic microbial metabolism is BOD<sub>5</sub>:N:P which is equal to 100:5:1 (von Sperling, 2014). Table 2 shows the N and P concentration data and the BOD<sub>5</sub>:N:P proportion for the few studies that quantified these three parameters. Except for effluents coming from the bleaching stage, as reported by Amaral et al. (2013), other samples need P supplementation, and all need N for good microbial activity in the bioreactor. These data support the need for supplementing these nutrients to remove biologically organic matter (Ashrafi et al., 2015).

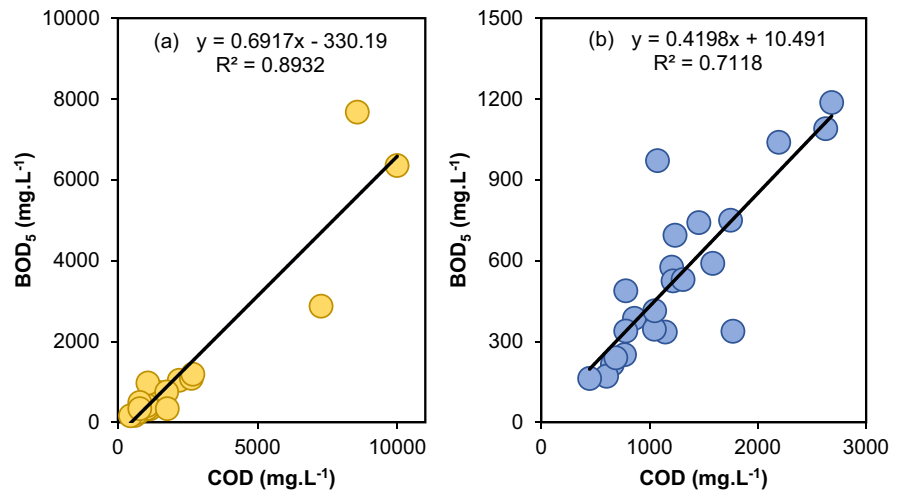
BOD<sub>5</sub> analysis takes at least 5 days (BOD<sub>5</sub>), while COD analysis takes 2 h (Dezotti, 2008). In this study, an important objective was to find a linear correlation between BOD<sub>5</sub> and COD values for pulp and paper industry effluents to facilitate characterizing and monitoring wastewater treatment based on the latter parameter, which can be obtained more quickly. The linear correlation coefficient is high if it is at least 0.8 and moderate if it is between 0.5 and 0.8 (Santos, 2007). Figure 2a shows the straight-line equation obtained from the data in Table 1 for raw effluents containing both reported parameters. The R<sup>2</sup> value was close to 0.9, a significant value. However, if the three most concentrated effluents coming from mechanical processes are excluded, and which are different from the others, the correlation coefficient decreased to a moderate value (Fig. 2b). Thus, it was not possible to obtain a prediction model using only effluents from the kraft process. Equations to estimate BOD<sub>5</sub> from COD with R<sup>2</sup> values above 0.8 have been obtained for domestic sewage in Brazilian cities (Budeiz & Aguiar, 2020; Silva & Mendonça, 2003). Some studies have also verified correlations between these and other

**Table 2** Proportion between organic matter (BOD<sub>5</sub>) and nutrients for raw effluents coming from pulp and paper industries

N (mg L <sup>-1</sup> )	P (mg L <sup>-1</sup> )	BOD <sub>5</sub> :N:P	Reference
6.5	27	100:1.1:4.6	Amaral et al. (2013)
2.7	11	100:0.4:1.5	
5.76	1	100:3.5:0.6	Brandão (2014)
5.4	0.897	100:0.5:0.1	Cabrera-Padilla and Pires (2014)
0.76	1.36	100:0.1:0.2	Rabelo et al. (2014)
0.66	1.37	100:0.1:0.2	
13.66	1.69	100:1.2:0.1	Magnanti et al. (2018)



**Fig. 2** COD×BOD<sub>5</sub> data for various raw effluents from pulp and paper industries. **a** All effluents; **b** all effluents except those from mechanical processes



physicochemical parameters for industrial effluents. Liang et al. (2018), for example, found a linear correlation between COD and toxicity for textile effluents in China, with  $R^2$  values above 0.82. Costa et al. (2022) reported a linear correlation between COD and BOD<sub>5</sub> for cassava processing wastewaters generated in Brazil. On the other hand, Ramos et al. (2020) obtained lower coefficients when analyzing COD and BOD<sub>5</sub> values for textile industries (0.6737), and textile laundry industry effluents (0.6055) from Brazil.

Linear correlations between other parameters were also tested and are shown in Table 3. Calculations were made with and without effluent data from mechanical pulping processes. The  $R^2$  value for COD and TC, for example, was above 0.9. When excluding one sample from mechanical processes, it decreased to a value below 0.8, showing that outliers can cause erroneous linear correlation. Thus, mechanical pulping effluents were disregarded for further attempts of linear correlation. Evaluating the BOD<sub>5</sub> data with AC and TC,  $R^2$  was 1.0. However, only two samples were used for obtaining such correlations. When analyzing COD with total dissolved solids (TDS),  $R^2$  value was above 0.8 (Fig. 3); for BOD<sub>5</sub>×TDS, the amount of data was very low. TDS is measured gravimetrically after evaporating the filtered sample (von Sperling, 2014). Compared with COD, TDS determination does not require the use of chemicals and it could be used to predict COD concentration in raw effluents emanating from chemical pulping processes. Other correlation attempts showed very low  $R^2$  values.

### Characteristics of secondary effluent

In addition to the raw effluent characteristics, many studies in the literature have presented data on secondary effluents (Table 4). Some samples were not in accordance with discharge standards. Thus, many of these studies intended to evaluate tertiary treatments. The composition of secondary effluents also showed sample variations. Kraft effluent samples predominated, while some studies made no mention of the type of pulping process employed.

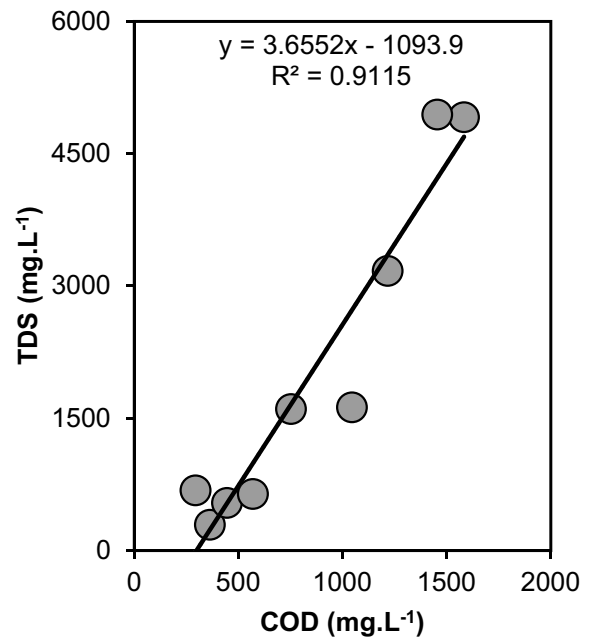
These effluents have pH values within the ranges established by CONAMA for disposal, between 5 and 9 (Brazil, 2011), since they had already undergone treatment stages requiring pH adjustments. These adjustments are required prior to biological treatments (von Sperling, 2014), which may explain the pH values being near 7. As was expected, it was observed that physicochemical parameter values for secondary effluents were lower than raw effluent values. It is important to mention that some secondary effluent COD values were higher than what were observed for raw effluent samples when comparing effluents from different industries. Comparing TSS, BOD<sub>5</sub>, and COD mean values for secondary effluents with state agency limits (Morais & Santos, 2019), one can clearly see that the first two values are lower, while COD is not, meaning that tertiary treatments are needed, or that conventional treatments need to be improved, although it is also important to know the depuration capacities of the receiving water bodies. For some samples, TP concentration was well above

**Table 3** Correlations between organic matter (COD or BOD<sub>5</sub>) and solids, color, turbidity, or total phenols for raw effluent data from the pulp and paper industries

Parameters	Equation	R <sup>2</sup>	No. of samples
Correlations for all data			
COD×BOD <sub>5</sub>	$y=0.6917x-330.2$	0.8932	26
COD×TS	$y=0.6888x+1630.9$	0.3989	13
COD×TSS	$y=0.1408x+206.3$	0.6648	18
COD×TDS	$y=0.4679x+1539.2$	0.2750	10
COD×TP	$y=-0.0453x+103.2$	0.4756	5
COD×turbidity	$y=0.0846x+127.4$	0.4379	11
COD×AC	$y=0.7200x+433.6$	0.3947	8
COD×TC	$y=1.1831x-749.4$	0.9674	6
BOD <sub>5</sub> ×TS	$y=1.0384x+2673.5$	0.2035	8
BOD <sub>5</sub> ×TSS	$y=0.1523x+405.1$	0.4747	12
BOD <sub>5</sub> ×TDS	$y=0.3115x+3414.7$	0.0553	5
BOD <sub>5</sub> ×TP	$y=-0.0425x+65.5$	0.0886	5
BOD <sub>5</sub> ×turbidity	$y=0.1527x-15.0$	0.7766	7
BOD <sub>5</sub> ×AC	$y=1.0927x-53.7$	1.0000	2
BOD <sub>5</sub> ×TC	$y=3.3056x-1737.1$	0.9934	3
Correlations without effluent data from mechanical processes			
COD×BOD <sub>5</sub>	$y=0.4198x+10.5$	0.7118	23
COD×TS	$y=3.4902x-778.6$	0.6616	12
COD×TSS	$y=0.0221x+316.1$	0.0027	15
COD×TDS	$y=3.6552x-1093.9$	0.9115	9
COD×TP	$y=-0.0453x+103.2$	0.4756	5
COD×turbidity	$y=-0.2549x+410.4$	0.1495	8
COD×AC	$y=0.7200x+433.6$	0.3947	8
COD×TC	$y=1.3180x+918.0$	0.7772	5
BOD <sub>5</sub> ×TS	$y=0.1495x+3154.3$	0.0003	7
BOD <sub>5</sub> ×TSS	$y=0.1533x+331.0$	0.0237	9
BOD <sub>5</sub> ×TDS	$y=8.8910x+1280.3$	0.8559	4
BOD <sub>5</sub> ×TP	$y=-0.0425x+65.5$	0.0886	5
BOD <sub>5</sub> ×turbidity	$y=0.0055x+53.1$	0.0001	4
BOD <sub>5</sub> ×AC	$y=1.0927x-53.7$	1.0000	2
BOD <sub>5</sub> ×TC	$y=-1.245x+1279.6$	1.0000	2

the limit value allowed by CONAMA (Brazil, 2011). These compounds are recalcitrant and are partly responsible for the dark color of the effluent and its toxicity (Hubbe et al., 2016).

TOC was also quantified in secondary effluents. It ranged from 54.2 to 250 mg L<sup>-1</sup> (Giroletti, 2017; Hinojosa & Xavier, 2016; Matias et al., 2018; Moreira, 2012; Rocha, 2017; Rodrigues, 2011). SSS were analyzed in only three studies, and all values were below the limit value of 1 mL L<sup>-1</sup> (Rezende

**Fig. 3** COD×TDS data for various raw effluents from pulp and paper industries, except those from mechanical processes

et al., 2010; Moreira, 2012; Farias & Ferreira, 2014). Sulfide was quantified in Farias and Ferreira (2014) and Magnanti et al. (2018), whose concentrations were below the limit value of 1 mg L<sup>-1</sup> for discharge (Brazil, 2011). Ammoniacal nitrogen was analyzed by four studies (Rezende et al., 2010; Moreira, 2012; Bonfim et al., 2013; Lopes, 2018). Values were below the 20 mg L<sup>-1</sup> limit set by federal legislation (Brazil, 2011). AOXs were also analyzed in a few samples, specifically for effluents from pulp bleaching steps (Rezende et al., 2010; Rodrigues, 2011; Moreira, 2012; Lopes et al., 2013). Chlorides were rarely quantified (Rezende et al., 2010; Azzolin & Fabro 2012; Moreira, 2012; Giroletti, 2017). Samples had AOX concentrations between 1.5 and 4.59 mg Cl L<sup>-1</sup> and chloride concentrations from 0 to 493 mg L<sup>-1</sup>.

When calculating the BOD<sub>5</sub>/COD ratio, it was found that secondary effluent values were generally lower than raw effluent values. This shows that treatments adopted in the industries reduce more biodegradable organic matter fractions. If effluents need to be polished to comply with the legislation, physicochemical processes are more indicated, since the BOD<sub>5</sub>/COD ratio in many samples was well below 0.4.

**Table 4** Secondary effluent characteristics from Brazilian pulp and paper industries

Process	pH	BOD <sub>5</sub> (mg L <sup>-1</sup> )	COD (mg L <sup>-1</sup> )	BOD <sub>5</sub> /COD	Color (mg PtCo L <sup>-1</sup> )	Turbidity (NTU)	Lignin	TP (mg L <sup>-1</sup> )	TS (mg L <sup>-1</sup> )	TSS (mg L <sup>-1</sup> )	References
Kraft with bleaching	–	–	187	–	288	–	–	–	–	–	Matias et al. (2018)
Kraft with bleaching	7.6	68	375	0.18	1369	–	–	–	2446	60	Rezende et al. (2010)
Kraft with bleaching	7.5–7.9	52	176	0.29	–	–	–	–	–	–	Rodrigues (2011)
Kraft with bleaching	–	17	196	0.09	489 (TC)	36	–	–	1455	48	Moreira (2012)
Kraft with bleaching	–	15	81	0.18	104 (TC)	19	–	–	1380	36	
Kraft with bleaching	–	55	490	0.11	816 (TC)	–	–	–	–	30	Araújo (2013)
Kraft with bleaching	–	13	76	0.17	409	37	–	–	–	–	Kossar et al. (2013)
Kraft with bleaching	–	45	263	0.17	794 (TC)	–	20 mg phenol L <sup>-1</sup>	–	–	–	Lopes et al. (2013)
Kraft with bleaching	8.3	–	180	–	–	–	–	–	–	–	Marques et al. (2013)
Kraft with bleaching	7.8	169	328	0.51	0.3 (420 nm) (AC)	–	1.6 (280 nm)	–	–	–	Rocha (2017)
Kraft with bleaching	6.7	–	360	–	575	<0.1	–	–	2000	10	Santos et al. (2020)
Kraft without bleaching	6.9–7.7	78	408	0.19	840 (TC)	320	2.9 (280 nm)	–	–	–	Hinojosa and Xavier (2016)
Kraft	–	32	425	0.075	1404	–	–	–	–	–	Rosa et al. (2013)
Kraft	7.8	2.2	150	0.014	294 (TC)/364 (AC)	6.2	–	–	1165	8	Quartaroli et al. (2014)
	7.5	18	558	0.033	2195 (TC)/2495 (AC)	56	–	–	1732	84	
	7.5	23	583	0.039	1784 (TC)/1884 (AC)	61.5	–	–	–	–	
Kraft	–	–	166	–	493 (TC)	17 mg L <sup>-1</sup>	11 mg L <sup>-1</sup> /1.69 (280 nm)	–	1107	26	Follmann (2017)
Kraft	–	143	341	0.42	139	1.6	0.799 (280 nm)	–	1265	29	Giroletti (2017)

Table 4 (continued)

Process	pH	BOD <sub>5</sub> (mg L <sup>-1</sup> )	COD (mg L <sup>-1</sup> )	BOD <sub>5</sub> /COD	Color (mg PtCo L <sup>-1</sup> )	Turbidity (NTU)	Lignin	TP (mg L <sup>-1</sup> )	TS (mg L <sup>-1</sup> )	TSS (mg L <sup>-1</sup> )	References
Kraft	7.9	–	948	–	2963 (TC)/4049 (AC)	221	95 mg L <sup>-1</sup>	–	1918	171	Neves et al. (2017)
Kraft	7.0	–	903–943	–	2190–2403 (AC)	190–224	–	9–14.3	–	–	Souza et al. (2017a)
Kraft	7.0	–	275	–	717 (TC)/950 (AC)	32	–	25	–	–	Souza et al. (2017b)
Kraft	7.7	10	132	0.08	544 (TC)	5.5	–	0.03	–	44	Lopes (2018)
Kraft	7.8	61	173	0.35	–	–	–	0.09	–	50	Magnanti et al. (2018)
Kraft	8.3	–	141	–	707 (TC)	–	–	7.8	–	–	Bender et al. (2019)
Without bleaching	7.9	91	439	0.21	780	85	–	–	–	–	Farias and Ferreira (2014)
Paper production	–	27	77	0.35	–	–	–	–	–	32	Brandão (2014)
Not informed	7.7	41	253	0.16	114 (TC)	17.6	–	–	241	196	Azzolini and Fabro (2012)
Not informed	7.3	–	253	–	606 (AC)	25.1	–	–	1083	34	Bonfim et al. (2013)
Not informed	7.6	–	261	–	552 (AC)	24.8	–	–	1083	34	Dias (2013)
Not informed	6.4	17	371	0.05	1160 (AC)	54.1	–	–	–	62	Sakurai et al. (2016)
Not informed	7.6	–	620	–	392 (TC)/4290 (AC)	288	–	–	–	223	Marques et al. (2017)
Not informed	6.8	–	180	–	3200	–	–	–	–	–	Tilha et al. (2019)
Not informed	6.8	96	140	0.68	–	18	–	–	20	–	Neves et al. (2020)
Not informed	8.0	–	194	–	352 (AC)	6.1	–	12.2	–	–	

PtCo platinum–cobalt unit, TC true color, AC apparent color, NTU nephelometric turbidity unit, TP total phenols

**Table 5** Efficiency of the conventional effluent treatments (in percentage removal) used by Brazilian pulp and paper mills

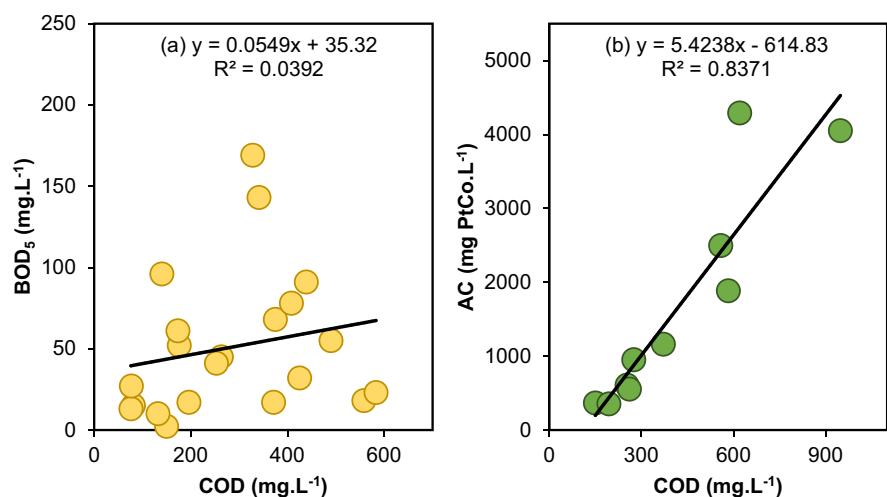
BOD <sub>5</sub> (%)	COD (%)	Color (%)	Turbidity (%)	TS (%)	TSS (%)	References
83.4	82.7	–	–	–	94.6	Brandão (2014)
73.1	75.2	– 160	– 97.7	–	–	Farias and Ferreira (2014)
58.6	67.4	50	61.9	38	93.1	Giroletti (2017)
94.9	93.5	–	–	–	87.1	Magnanti et al. (2018)
–	86	– 39.1	–	–	–	Matias et al. (2018)
71.7	82.1	–	85	99.4	–	Tilha et al. (2019)

To evaluate the efficiency of the conventional treatments, the percentage removal of some parameters was calculated as shown in Table 5. It was noted that COD and BOD<sub>5</sub> removals varied from 67 to 94% and from 59 to 95%, respectively. According to CONAMA, the minimal BOD<sub>5</sub> removal is 60% (Brazil, 2011). Except Giroletti (2017), all studies are in accordance with this discharge standard. TSS removal was high, above 87%. In opposite to Giroletti (2017), color has increased after biological treatment according to two studies. This increase can be attributed to the partial biodegradation of wood-derived pollutants into more colored compounds (Peitz & Xavier, 2017). Despite color being a parameter without a limit value allowed by CONAMA, which only states that the effluent should not alter the natural color of the watercourse (Brazil, 2011), this parameter is very important with regard to the discharge of treated pulp and paper mill effluents. The linear correlation between all COD and BOD<sub>5</sub> data for secondary effluents was also evaluated here (Fig. 4a). The  $R^2$  value was much lower than 0.8, making it impossible to estimate BOD<sub>5</sub> from the COD for secondary effluents. Ferreira et al. (2013) evaluated

correlations between COD and color (apparent and true colors) for effluents from a pulp and paper industry. They used raw, secondary, and tertiary effluent data. The only significant coefficient ( $R^2 > 0.9$ ) was for COD and apparent color data for secondary effluents. They suggested the replacement of COD for apparent color to reduce costs and waste from laboratory analysis. Several attempts at the linear correlation in our study involving other characteristics were also made, as shown in Table 6. Interestingly,  $R^2$  values were above 0.8 only for COD and apparent color (Fig. 4b), suggesting that the latter parameter can be used to predict the former. It is important to highlight that this correlation was obtained here for effluents from different industries. For the other attempts, the values were well below 0.8. Considering that COD data are used to monitor an effluent treatment plant, as suggested by Ferreira et al. (2013), an apparent color analysis is faster and simple and it could be used to predict COD in biologically treated effluents.

From the data in Tables 1 and 4, the ranges, averages, and standard deviations of the analyzed parameters were determined, as shown in Table 7. The

**Fig. 4** a COD × BOD<sub>5</sub> and b COD × apparent color data for various biologically treated effluents from pulp and paper industries



**Table 6** Correlations between organic matter (COD or BOD<sub>5</sub>) and solids, color, turbidity, or total phenols for secondary effluent data from pulp and paper industries

Parameters	Equation	R <sup>2</sup>	No. of samples
COD×BOD <sub>5</sub>	$y = 0.0549x + 35.3$	0.0392	21
COD×TS	$y = 1.4773x + 835.7$	0.2570	13
COD×TSS	$y = 0.1723x + 9.8$	0.3581	18
COD×TDS	$y = 1.0036x + 1015.2$	0.1597	12
COD×TP	$y = 0.1612x - 20.5$	0.7936	5
COD×turbidity	$y = 0.2802x - 27.3$	0.4101	20
COD×AC	$y = 5.4238x - 614.8$	0.8371	10
COD×TC	$y = 2.6924x - 61.8$	0.6620	15
BOD <sub>5</sub> ×TS	$y = -2.8134x + 1353.7$	0.0313	8
BOD <sub>5</sub> ×TSS	$y = -0.0374x + 58.1$	0.0009	12
BOD <sub>5</sub> ×TDS	$y = 1.1354x + 1268.2$	0.0064	7
BOD <sub>5</sub> ×TP	$y = 0.0012x + 0.0182$	1.0000	2
BOD <sub>5</sub> ×turbidity	$y = 0.3897x + 38.3$	0.0421	13
BOD <sub>5</sub> ×AC	$y = 82.7x + 230.7$	0.6485	4
BOD <sub>5</sub> ×TC	$y = 0.9411x + 768.8$	0.0010	10

literature on effluents from pulp and paper industries reported pH levels varying from 6.3 to 6.8 (Toczyłowska-Mamińska, 2017), which is a very narrow range compared to the data shown here. The same review states that COD can range from 500 to 115,000 mg L<sup>-1</sup>. Compared to the range in the present study, one can see that the lower limit is similar, while the upper limit is higher than that of effluents generated by industries in Brazil.

In addition to the aforementioned parameters, the temperature is a characteristic legislated by CONAMA (Brazil, 2011). This parameter was not widely reported in the consulted studies. Temperature ranges and averages were established with the data obtained for both effluent types. It was observed that all secondary effluents, in terms of temperature, were within the disposal limit, 40 °C (Brazil, 2011).

### Toxicity and estrogenic activity in pulp and paper mill effluents

Another relevant characteristic of pulp and paper industry wastewater is toxicity. Long-chain fatty acids, organochlorine compounds, and dioxins are present in this type of effluent and contribute most to its high toxicity (Hubbe et al., 2016; Lindholm-Lehto

**Table 7** Ranges, average, and number of samples for each raw and secondary effluent parameter taken from Brazilian pulp and paper industry data

Parameters	Range	Average	No. of samples
Raw effluent from Brazilian industries			
pH	3–11.2	–	17
BOD <sub>5</sub> (mg O <sub>2</sub> L <sup>-1</sup> )	163–7674	1125.9 ± 1824.3	26
COD (mg O <sub>2</sub> L <sup>-1</sup> )	295–9992	1836.3 ± 2227.0	35
BOD <sub>5</sub> /COD	0.19–0.9	0.45 ± 0.17	26
AC (mg PtCo L <sup>-1</sup> )	475–3124	1204.8 ± 868.8	8
TC (mg PtCo L <sup>-1</sup> )	46–7780	2012.7 ± 3048.3	6
Turbidity (NTU)	4.2–1439	374.5 ± 475.8	11
TP (mg L <sup>-1</sup> )	0.07–84	32.2 ± 43.7	5
TS (mg L <sup>-1</sup> )	590–5969	2578.9 ± 1979.2	13
TSS (mg L <sup>-1</sup> )	20–1771	529.8 ± 518.2	18
TDS (mg L <sup>-1</sup> )	292–4942	2044.3 ± 1851.8	9
TSS/TS	0.03–0.51	0.22 ± 0.20	9
Temperature (°C)	20–50	34.9 ± 11.4	5
Secondary effluent from Brazilian industries			
pH	6.4–8.3	–	26
BOD <sub>5</sub> (mg O <sub>2</sub> L <sup>-1</sup> )	2.2–169	51.1 ± 44.3	21
COD (mg O <sub>2</sub> L <sup>-1</sup> )	76–948	297.0 ± 188.6	33
BOD <sub>5</sub> /COD	0.014–0.68	0.21 ± 0.17	21
AC (mg PtCo L <sup>-1</sup> )	352–4290	1670.2 ± 1670.2	10
TC (mg PtCo L <sup>-1</sup> )	104–2963	883.1 ± 808.0	15
Turbidity (NTU)	0.1–320	65.6 ± 94.9	20
TP (mg L <sup>-1</sup> )	0.03–25	9.0 ± 10.3	5
TS (mg L <sup>-1</sup> )	20–2446	1241.2 ± 659.3	12
TSS (mg L <sup>-1</sup> )	8–223	65.4 ± 63.6	18
TDS (mg L <sup>-1</sup> )	45–2386	1344.9 ± 583.03	12
TSS/TS	0.01–0.81	0.10 ± 0.23	12
Temperature (°C)	17.2–37	25.8 ± 7.5	12

et al., 2015). This is not only worrying for disposal of treated effluent, but also the biological treatment of effluent since certain compounds harm microorganisms involved in this stage (Hubbe et al., 2016). It is known that effluents emanating from each process stage have distinct toxicities, and effluents generated from the bleaching process are more toxic (Furley et al., 2015, 2018). More recently, toxic substance concentrations in bleaching effluents have been reduced because chlorine gas (Cl<sub>2</sub>) has not been used as an input (Hubbe et al., 2016), in addition to other improvements in the productive process (Furley et al., 2018). ClO<sub>2</sub> was an alternative substitute for Cl<sub>2</sub>, which is less harmful (Cabrera, 2017). Nonetheless,

wastewater from this stage is still toxic and was reported by some studies.

Regarding the toxicity data, Maria et al. (2014) studied two raw kraft pulp bleaching effluents, one alkaline and the other acidic. Both the individual and mixed samples showed high chronic and acute toxicity for *Daphnia similis* and *Ceriodaphnia dubia*, respectively. The alkaline effluent was more toxic according to the authors. Using the same bioindicators, Chaparro et al. (2010a, b) and Chaparro and Pires (2011) also observed chronic and acute toxicities in mixed effluents (60% v/v alkaline, 40% v/v acidic effluent) from kraft pulp bleaching. This mixture of effluents also showed cytotoxic, genotoxic, and mutagenic effects on *Allium cepa* L. (Chaparro & Pires, 2011). For *Daphnia magna*, a kraft process effluent showed acute toxicity (Micheletto et al., 2019). On the other hand, the effluent evaluated by Giroletti (2017) was not toxic for lettuce seeds (*Lactuca sativa*). According to the author, the company where the effluent was collected does not bleach the pulps; therefore, the lack of chlorinated residue led to no toxicity. Using *D. magna* as a test organism, other studies have also verified no toxicity for non-bleached kraft pulp effluents (Castro et al., 2019; Machado, 2017).

Secondary effluent toxicity has also been reported in some of the reviewed studies. Lopes et al. (2013) observed that bleached kraft pulp effluent did not present acute toxicity to *D. similis*, but it showed chronic toxicity for *Pseudokirchneriella subcapitata*, since it inhibited its growth, and reduced the survival and reproduction of *C. dubia*. According to the authors, organic matter, lignin, and AOX contributed to these results. However, when compared to a primary effluent from the same company, the secondary effluent was less toxic. It should be highlighted that two other studies showed no toxicity for secondary effluent samples using three tested organisms: *D. similis*, *C. dubia*, and *Raphanus sativus* (radish) seeds (Brandão, 2014; Lopes et al., 2020). The secondary effluent studied by Neves et al. (2020) promoted higher inhibition on *L. sativa* when effluent concentration was increased from 0.1 to 100%. Additionally, necrosis was noted in 20% of the seeds exposed to 100% effluent.

Estrogenic activity is another important parameter to evaluate in these effluents since it results in abnormalities in the reproductive system of animals

(Furley et al., 2018; van den Heuvel, 2010). When analyzing raw effluents from three countries, Canada, Brazil, and New Zealand, it was found that effluent from Brazil had higher estrogenic activity for *Oncorhynchus mykiss* (Orrego et al., 2017). According to the authors, estrogenic effects on this fish were related to the type of pulping process (kraft > elementary chlorine-free kraft > TMP). Castro et al. (2018b) observed that 14-day exposure with 4% raw effluent from a bleached kraft pulp company affected the hepatic and testicular physiology and biochemistry in zebrafish (*Danio rerio*). For an unbleached kraft pulp raw effluent, Alves et al. (2014) observed estrogenic activity for recombinant *Saccharomyces cerevisiae*. For a secondary effluent (from bleached kraft pulp), Lopes et al. (2013) observed estrogenic activity for a similar organism test. This study also compared the primary effluent from the same company, concluding that the activated sludge treatment decreased, but did not eliminate these estrogenic effects. In turn, Oliveira et al. (2020) exposed immature rat testis and Sertoli cells to 4% secondary effluent for 1 h, verifying alterations in energy metabolism and mitochondrial activity, respectively. Together, these data show that conventionally treated effluents from Brazilian pulp and paper industries can still contain toxic and estrogenic compounds.

### Conventional treatments used in Brazilian mills

Table 8 shows another important aspect of this data survey, referring to company unit operations of this sector to treat their effluents. Most use grates, followed by a primary decanter, and activated sludge biological treatment (Fig. 5). Interestingly, preliminary treatments are rarely mentioned in literature reviews (Ashrafi et al., 2015; Cabrera, 2017; Hubbe et al., 2016). A big problem in primary and secondary decanters is the considerable amount of generated sludge. To minimize this problem, Stoppa et al. (2014) found that adding certain substances to the bioreactor, like folic acid (0.5 mg L<sup>-1</sup>), reduced sludge formation by 24%, but did not reduce COD. However, the chronic toxicity of the treated effluent was increased. Generated sludge can also be used as a soil conditioner, in compost, civil construction, or to generate biofuels and renewable energy via fermentation or thermochemical processes

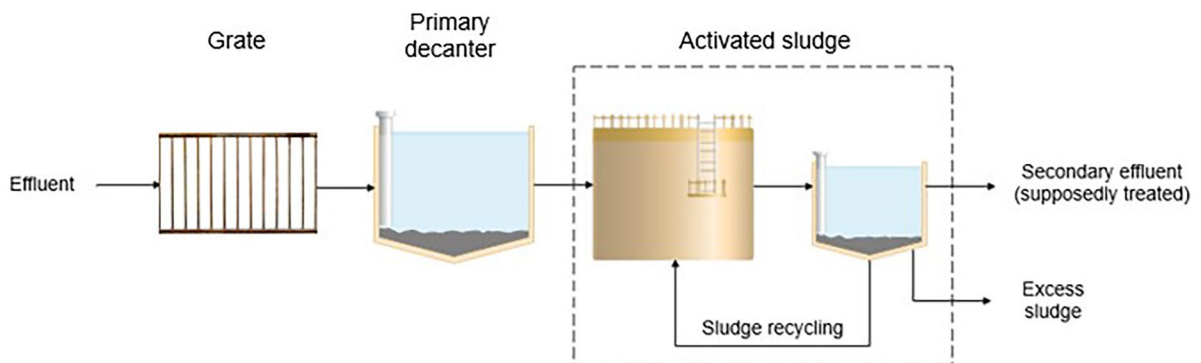


**Table 8** Types of treatment used in the pulp and paper industries in Brazil

Treatments	Reference
Grating, neutralization, primary decanter, cooling, activated sludge	Rodrigues (2011)
Grating, neutralization, 2 primary decanters in series, cooling, 2 aeration tanks in parallel with N and P supplementation, 4 secondary decanters in parallel (activated sludge system)	Moreira (2012)
Primary settling pond, aerated pond, polishing pond	Vanzetto (2012); Machado (2017)
Neutralization and homogenization, primary treatment, aeration, activated sludge with pure oxygen, coagulation–flocculation–decantation	Ferreira et al. (2013)
Grating, sieving, primary decanter, cooling, 2 aeration tanks in series, 2 secondary decanters in parallel (activated sludge system), coagulation–flocculation–ultrafiltration	Kossar et al. (2013)
Desanding, grating, primary settling pond, facultative aerated pond, secondary settling pond	Rosa et al. (2013)
Primary decanter, aerated pond, secondary settling pond	Brandão (2014)
Grating, primary decanter, supplementation, activated sludge, coagulation–flocculation–decantation	Farias and Ferreira (2014)
Grating, desanding, primary decanter, supplementation, pH correction, cooling, MBBR, 2 aeration tanks in series, 2 secondary decanters in parallel (activated sludge)	Quartaroli et al. (2014); Sakurai et al. (2016); Follmann (2017)
Grating, equalization, primary decanter, 2 aeration tanks in series, secondary decanter (activated sludge)	Giroletti (2017)
Primary decanter, activated sludge system	Lopes (2018)
Flotation or primary decanter, aeration tank, 2 secondary decanters in parallel (activated sludge), polishing ponds	Magnanti et al. (2018)
Grating, primary decanter, activated sludge	Bender et al. (2019)
Sieving, coagulation–flocculation–decantation, activated sludge	Caldeira (2019)
Grating, desanding, sieving, flotation, homogenization, cooling tower, activated sludge	Tilha et al. (2019)
Primary decanter, activated sludge	Vidal et al. (2019)
Sieving, primary decanter, aeration pool, neutralization, settling, activated sludge, polishing pond	Neves et al. (2020)
Primary decanter, activated sludge system	Santos et al. (2020)

(incineration, pyrolysis) (Lopes et al., 2018; Simão et al., 2018). Primary treatment through flotation was also verified. Some companies use coagulation–flocculation, which can be applied as a primary treatment (Caldeira, 2019) or polishing (Farias

& Ferreira, 2014; Ferreira et al., 2013). However, chemical sludge from coagulation–flocculation is more problematic because it is not biodegradable and generally must be disposed of in landfills or must be incinerated (Simão et al., 2018).

**Fig. 5** Effluent treatment system adopted in most Brazilian pulp and paper industries

Other biological treatment processes, beyond activated sludge, which is used by Brazilian industries, are aerated ponds, moving bed biofilm reactors (MBBR), and polishing ponds. These treatments are aerobic and have been widely reported in the literature (Ashrafi et al., 2015). Polishing ponds have been used for improving the activated sludge system (Magnanti et al., 2018; Neves et al., 2020). No evidence for anaerobic biological processes was found in the consulted studies.

For four of the 18 companies mentioned, the neutralization step before biological treatment, which requires pH levels close to 7, was mentioned (von Sperling, 2014). Supplementation has been used to supply N and P; however, both neutralization and supplementation are common procedures for treating effluents at many companies in the sector (Ashrafi et al., 2015; Cabrera, 2017). It is important to mention that effluent treatment from these industries does not seek to remove N and P, even when both are supplemented. Thus, supplementation must be controlled to avoid eutrophication into receiving water bodies (Ashrafi et al., 2015).

It should be highlighted that most studies characterized these effluents to alternatively and more effectively treat them. Conventional processes generally comply with legislation, but have limitations, especially regarding a lot of excess sludge, which must be disposed of in landfill or incinerated (Simão et al., 2018), in addition to secondary effluent non-compliance with current legislation in some cases. Regarding using treatments that generate less sludge, anaerobic biological processes have been tested and can produce combustible biogas (Gao et al., 2016; Veroneze et al., 2019). Additionally, the requirement by power and nutrients is lower than that in the case of aerobic processes. If this gas is used properly, it could be burned to supply a part of the industry's energy demands (Ashrafi et al., 2015). Interestingly, biogas production from treatment of industrial effluents has increased in Brazil (Ramos et al., 2022). Anaerobic digestion can also be used to treat primary sludge and, mainly, secondary sludge (Bayr & Rintala, 2012; Karlsson et al., 2011; Lopes et al., 2018). Thus, these wastes can be converted into biogas, saving energy to the process. Furthermore, many other alternative forms of treatment are effective (Hubbe et al., 2016; Kamali & Khodaparast, 2015), but technical and economic challenges still need to be overcome. Anyway, this review can help

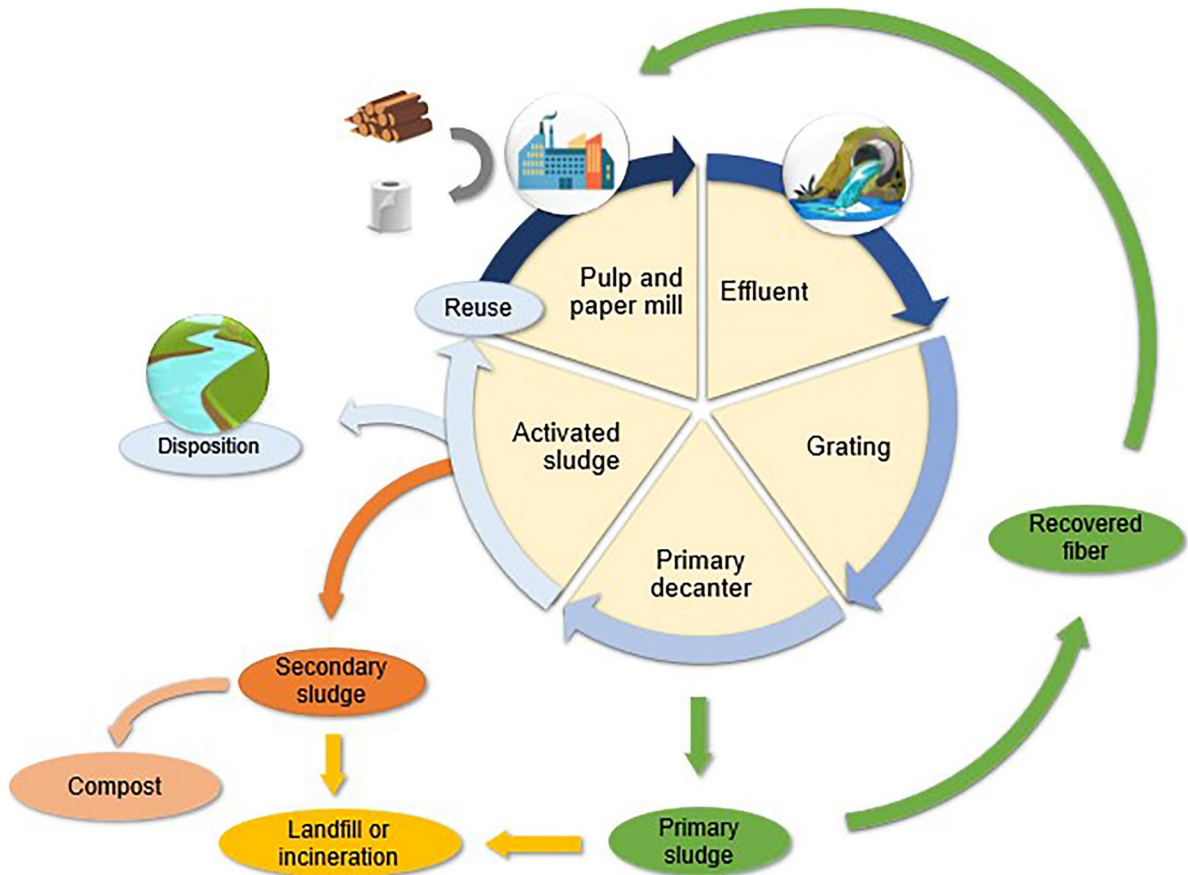
in improving conventional treatments or choosing new technologies to be tested to treat more appropriately these effluents.

### Reuse of treated effluents

Reusing treated effluent is an important aspect for reducing huge water consumption in the pulp and paper industries. As a result, discharged effluent to the environment is also reduced, leading to economic and sustainable benefits for less water use (Cabrera, 2017; Li et al., 2016; Mandeep et al., 2019). Some process steps need freshwater, while others require low-quality water (Li et al., 2016). Analyzing an industrial wastewater treatment plant, Kossar et al. (2013) suggested reusing ultrafiltration-polished effluent in kraft paper machine showers and vacuum pumps, because the water does not enter into contact with the produced paper. The authors highlighted that the color of the treated effluent was the limiting factor for its reuse. A more recent study reported that three Brazilian industries reuse their effluents after treating them, and reuse levels ranged from 40 to 85% of all generated effluents. One of the evaluated companies uses treated effluent to wash equipment and floors, for gardening purposes, to prepare glues, to separate shavings, and to cool hot water (Monteiro et al., 2018).

For irrigation purposes, Rezende et al. (2010) reported that treated effluent can be used to irrigate eucalyptus plantations, which is the main raw material used in Brazilian pulp and paper mills. In turn, Almeida et al. (2017) applied treated effluent on soil for 6 years, a period of a complete rotation of eucalyptus cultivation. It was noted that irrigation did not affect soil strength, hydraulic conductivity, and Ca, Mg, and organic carbon contents, while reducing mean aggregate size and increasing clay dispersion and pH. The eucalyptus growth was not harmed. However, higher values of Na adsorption rate and intermediate electrical conductivity were observed, indicating that soil quality can be impaired over a longer period of irrigation with treated effluent, requiring monitoring over the years.

The main aspects related to effluent treatment in this review are shown in Fig. 6, including reuse. It is important to highlight that the cycling of wastes in pulp and paper mills is within the circular economy context (Molina-Sánchez et al., 2018). The



**Fig. 6** Possible fate of treated effluent and sludge generated in pulp and paper mills

fibers recovered in the primary treatment can be returned to the mill or commercialized, reducing the generation of solid waste (Tilha et al., 2019). Using treated effluent in the industrial processes (Monteiro et al., 2018) or for irrigation (Rezende et al., 2010) and using residual sludge in agriculture (Simão et al., 2018) can reduce the water consumption and provide agricultural inputs in planting trees which are sources of vegetable fibers.

## Conclusion

This literature review presented the physicochemical parameters for raw effluents from Brazilian pulp and paper industries, demonstrating that they are quite diversified in terms of composition due to the different types of process employed (pulping,

bleaching), the different papers produced, and the different raw materials and inputs used. It was confirmed that raw effluent, in general, does not comply with Brazilian federal and state legislation for discharge, while secondary effluent partially complies with legislation. When analyzing the  $BOD_5/COD$  ratio, 65% of the raw effluent samples showed values above 0.4, meaning they can be biologically treated. On the other hand, some secondary effluent samples showed ratios well below 0.4, meaning that polishing through physicochemical treatments would be better if required to comply with disposal standards. The linear correlation coefficients for COD and  $BOD_5$  data were below 0.8, for both raw and secondary effluents, which means that  $BOD_5$  cannot be estimated from the COD values. On the other hand, strong correlations were observed between COD and dissolved solids for raw effluents ( $R^2=0.9115$ )

and between COD and apparent color for secondary effluents ( $R^2=0.8371$ ). It appears that, despite variations in effluent composition among Brazilian industries, or within the same company, the treatments are similar, consisting of grating, primary decanter, and activated sludge biological process. Due to the low content of N and P, it was verified that supplementing nutrients is common to biologically treat these effluents. The increase of the color after biological treatment, in addition to the total phenols and COD values which do not meet the legislation standards, is the main problem faced when treating these effluents. Improvements are suggested in existing biological treatment systems or the adoption of polishing steps to reduce/eliminate toxicity and estrogenic activity, which are still present in conventionally treated effluents.

**Author contribution** Márcio D. N. Ramos, Angélica S. Rangel, Karina S. Azevedo, Maria Gabriela B. Melo, Maria Caroline Oliveira, Camila M. U. Watanabe, and Fernanda F. Pereira: conceptualization, data curation, formal analysis, and writing of the original draft; Claudio Mudadu Silva: conceptualization and writing including review; André Aguiar: conceptualization, writing (including review), project administration, supervision, and visualization.

**Funding** This study was financed in part by the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES), Finance Code 001. Márcio D. N. Ramos received the student fellowships from the Fundação de Amparo à Pesquisa do Estado de Minas Gerais (Fapemig) and Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq). The English grammar review of this article was funded by the Natural Resources Institute of the Federal University of Itajubá.

**Data availability** The authors confirm that the data supporting the findings of this study are available within the article.

## Declarations

**Ethics approval and consent to participate** Not applicable.

**Consent for publication** Not applicable.

**Conflict of interest** The authors declare no competing interests.

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