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***Review Article** (Open Access)*

PAPER INDUSTRIES CONCERN WATER POLLUTION: A REVIEW

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ABSTRACT
The review attempted to present the scenario of the growing-up paper industries and their impacts on the environment. The study considered the data and information published in Journals, newspapers, and reports from different government and non-government organizations from 1970 to 2020. The review observed that the paper industries discharged effluent worldwide approximately 69094 million m^3/year. The paper industrial effluents contained higher BOD, COD, EC, and TSS combined with several chemicals like heavy metals, lignin, and organochlorine compounds that contaminated the surrounding water bodies and caused harm to aquatic life as well as human health. Hence, it should be treated before discharging into water bodies.

Keywords: *Effluent, Industry, Lignin, Paper, Pulp, Toxic*

INTRODUCTION
Paper, existing for about two thousand years, is competing successfully with modern information and communication technology (ICT) and many synthetic materials. Paper production all over the world is increasing rapidly. The Total world average per capita paper consumption was 49 kg in 1995 and 53 kg in 2018 (EU, 2015; FAO, 2020). The raw materials used in the pulp and paper industry are wood, straw, reeds, esparto grass, jute, flax, sisal, waste paper, etc.
Industrial processes used a large volume of water, and almost all of the water is released as effluent into the water environment. The discharged untreated or poorly treated industrial effluents on land and surface water bodies could relocate a huge cost to the environment (Rafique and Mostafa, 2020; Khan and Mostafa, 2011). The large volumes of effluent discharging industries are tannery industries, pulp and paper mills, textiles, sugar mills, thermal power plants, oil refineries, fertilizer industries (Islam and Mostafa, 2021). The Paper mill's water consumption rate is 230 to 500 m3 per ton of paper production to get fiber suspension from the raw materials (Waghmare, 1986). Hence, the pulp and paper industry is one of the vital industries that produced large amounts of hazardous effluents and are continuously releasing into surface water bodies.

GROWING-UP PAPER INDUSTRY
A strong correlation was observed between the increasing trend of paper consumption rate and the growth of gross national product (GNP). In Bangladesh, the per capita paper and board consumption rate was 5 kg, and in advanced countries, this rate is more than 200 kg, while the average rate was about 53 kg in 2018 (FAO, 2020) (Figure 1). The total paper consumption rate is increasing in Bangladesh and other developing countries.
Worldwide paper and board production was 408.84 million ton in 2018 and 399.79 million ton in 2010 (FAO, 2020). FAO, 2020 reported that Asia, North America, and Europe are the major paper producing regions (Figure 2). Almost 47 percent of the world's paper and board production is from Asia.
Figure 3 illustrates that the USA, Canada, Western Europe, and Japan produced more than 85% of the world's paper production in 1970. Now paper production in North America, Europe, and Oceania was decreasing, while the percentage was rising in many developing countries around the world especially in

Asia and Latin America (FAO, 2020; FAO, 2012; FAO, 1992; FAO, 1972; EU, 2015; EPN, 2018). Now Asia produces nearly 50% of pulp and paper globally, increasing from only 15% in 1970. China alone produces more than 25% of the world's paper production (FAO, 2018).

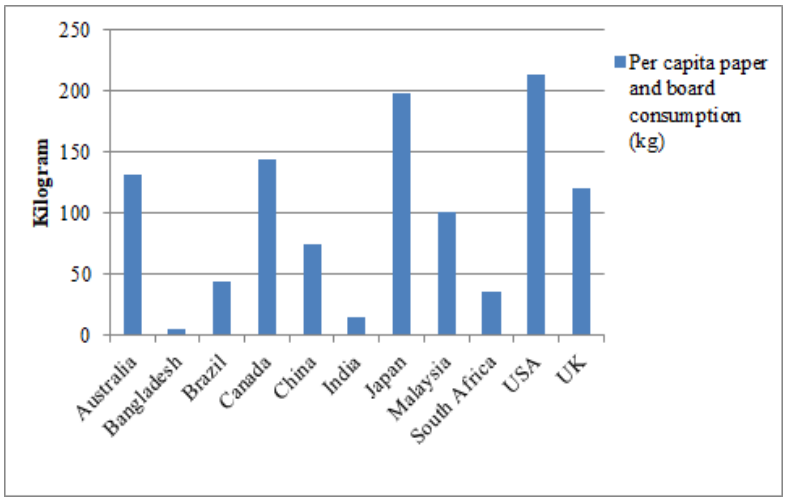

Figure 1: Per capita paper and board consumption in 2018 (FAO, 2020)

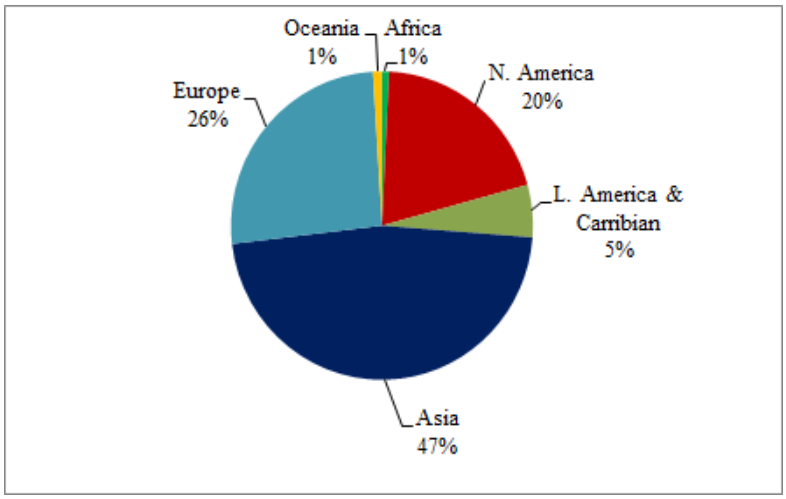

Figure 2: Paper production (%) by region in 2018 (FAO, 2020).

The growing-up pulp and paper industry requires a large amount of water. Wastewater generation, air emission, solid waste management, and energy consumption are the main environmental issues of the pulp and paper industry.

PAPER MANUFACTURING PROCESS

The raw materials of paper are cellulose, wood, pulp, or recycled paper (Bobu and Gavrilescu, 2010). Cellulosic fibers originated from non-wood raw materials such cereal straw, reeds, esparto grass, jute, flax, and sisal are the other sources of pulping raw materials (Gavrilescu *et al.*, 2009; González-García *et al.*, 2010; Puitel *et al.*, 2011; Rodríguez *et al.*, 2010). Raw materials are mixed with water in a paper mill to produce fiber suspension. Effluents contain large amounts of fiber, filler, and chemicals (surfactants, bleaching agents, glues, coloring agents, and biocides) that make it hazardous and toxic (Lacorte, 2003).

The main steps of the paper manufacturing process are raw material preparation, digestion, pulping, and bleaching (Figure 4). Raw material preparation, pulping, washing, screening, bleaching, and coating are the main pollution sources of pulp and paper mill. All the production stage process water has toxic components with different characterizations.

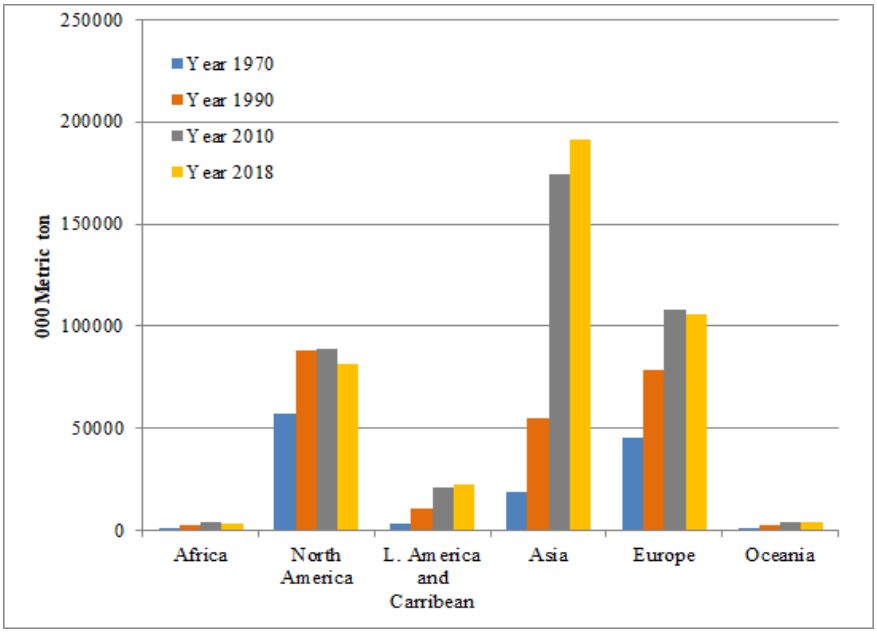

Figure 3: Paper and paperboard annual production by region.

Pulping is the vital constituent of paper production. Pulp properties depend upon the pulping process. Different types of pulping processes are used for getting a variety of quality of the paper. The pulping processes are mainly divided into four types: mechanical, chemical, recycled fiber, and textile fiber pulp (Table 1). Further, these types can be divided into subgroups. Every group and sub-group has a distinctive use of chemicals. A lot of chemicals are used in these processes.

About 3000 different chemicals are using in the paper production process, and about 200 of the most common chemicals are used. Usually, sodium salts are used for pulp processing, chlorine, chloride, and hydrogen peroxide are used in the bleaching process, sulfate of alumina, alkyl ketene dimer, abietic acid, sodium abietate are used for sizing in the paper industry. The chemicals, including zinc oxide, zinc sulfide for pigments, barium carbonate, casein, and others are used for deinking, additives, filler, dyeing agents (Worldofchemicals, 2020; paper web, 2020).

CHARACTERISTICS AND POLLUTION SOURCES

Cheremisinoff and Rosenfeld, (2010) reported that the pulp and paper industry discharge 100 million kg of toxic pollutants every year. Water consumption per ton of pulp production was approximately 200 m^3, and most of them are highly polluted, particularly wastewater generated from the chemical pulping process (Cecen *et al.*, 1992). Water consumption and effluent discharge from pulp and paper mills vary due to the different production processes, raw materials, and categories of paper. Table 2 illustrates that the average water consumption rate for per ton paper production was 256 m^3 while the average effluent discharge rate for per ton paper production was 169 m^3.

Worldwide paper and board production was 408.84 million ton in 2018 (FAO, 2020). Therefore, in the pulp and paper industry, worldwide freshwater consumption volume was 104663 million m^3, and wastewater discharge volume was 69094 million m^3 in 2018.

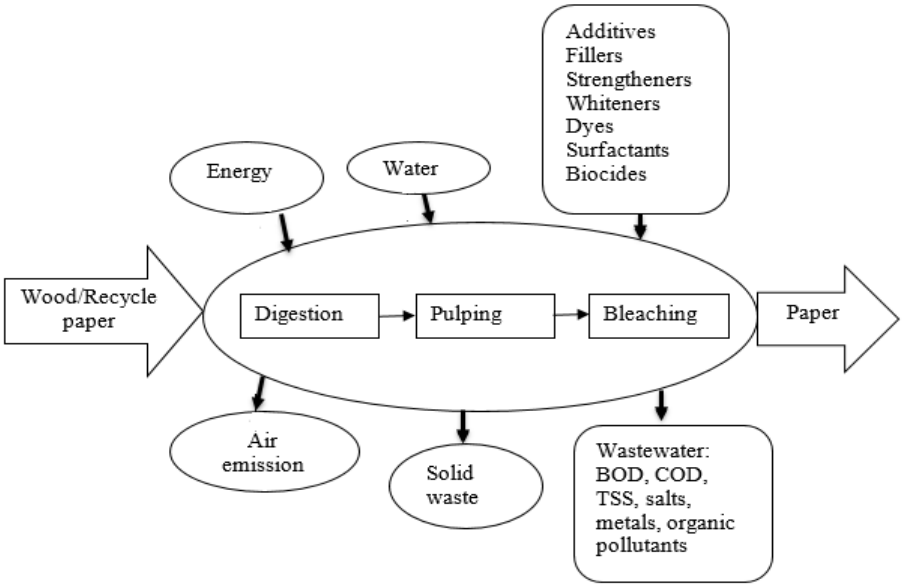

Figure 4: Schematic illustration of paper manufacturing process and pollutants.

Table 1: Pulping process, process chemicals and their purposes (KEMI, 2019; Singh, 2019)

Type of pulp	Pulp sub-groups	Identified chemicals and their purposes	Pulp properties
Mechanical pulp	Thermo-mechanical pulp (TMP)	Na_2SO_3 (if softwood) – Dissolve lignin NaOH or H_2O_2 (if hardwood) – Dissolve lignin and bleach pulp	Lignin content is higher. Short, weak, unstable fibers and light color.
	Chemi-Thermo-mechanical pulp (CMTP)	Pre-chemical impregnation with strong or weak alkali Na_2SO_3 (if softwood) – Dissolve lignin NaOH or H_2O_2 (if hardwood) – Dissolve lignin and bleach pulp	Lighter color and better strength properties than thermomechanical pulp
	Stone groundwood pulp (SGW)	Na_2SO_3 (if softwood) – Dissolve lignin NaOH or H_2O_2 (if hardwood) – Dissolve lignin and bleach pulp	Relatively short fibers
Chemical pulp	Sulphate pulp (Kraft pulp)	NaOH and Na_2S (white liquor) – Dissolve lignin Chlorine dioxide, oxygen, hydrogen peroxide, ozone – Bleach pulp	Pulp with low lignin content. Long, strong and stable fibers.
	Sulfite pulp	Bisulfites – Dissolve lignin	
Recycled pulp	Recycled pulp	NaOH – Deink pulp Soap – Deink pulp Fatty acids - Deink pulp H_2O_2 – Bleach pulp Chelating agents – Improve process efficiency e.g. bleaching	Mixture of fiber grades; properties depend on waste paper source
Textile fiber pulp	Textile fiber pulp	-	-

Table 2: Water consumption and effluent discharge from pulp and paper mills

Water consumption (m^3/ton paper production)	Average water consumption (m^3/ton paper production)	Effluent discharge (m^3/ton paper production)	Average effluent discharge (m^3/ton paper production)	Reference
200-240		180-220		Ramana, 1991
273-455		300		Subrahmanyam, 1990
-		10-50		Lacorte *et al.*, 2003
	256		169	
-		20-100		Gavrilescu et al.,2008
360-425		250-360		Kumer *et al.*, 1995
230-500		-		Waghmare, 1986
135		115		Karthik *et al.*, 2011
60		-		Lindholm-Lehto, 2015
-		100-250		Zazouli *et al.*, 2017

The pulp and paper industries pollute the environment due to the discharge of large volumes of wastewater, which contains various types of metallic, non-metallic, and toxic organic compounds (Table 3). The effluent physicochemical characteristics vary because of the different pulping, bleaching processes, and additives. But from every literature review, we observed that many physicochemical parameters exceed the permissible limit. The most alarming parameters of paper industries wastewater are BOD, COD, EC, TSS, heavy metals, lignin, and chlorophenol. Kumara et al. (2011) revealed that effluent from pulp and paper mills has high BOD, COD, lignin compound, and their derivatives, which were toxic and mutagenic to aquatic organisms. Moreover, this effluent contains some non-biodegradable compounds that can bioaccumulate in the water environment through the food chain. Heavy metals discharged from pulp and paper industry wastewater enters into the food chain can cause many health hazards like cancer, cardiovascular disease, nervous system disorder, kidney disease, respiratory disease, anemia, etc. (Mahurpawar, 2015; Fazeli, 1991; Nanda, 2014). Lead (Pb) released from kraft pulping damages the neurologic, hematologic, and renal systems in humans (Campana *et al.*, 2003). Cadmium (Cd) in the food chain causes toxicity to plants, animals, many microorganisms as well as kidney and bone damage of the human body (Mahurpawar, 2015). Calcium (Ca) and Magnesium (Mg) released from the pulping stage enact Neurotoxicity and toxicity to juvenile channel catfish (*Ictalurus punctatus*) (Gamelin *et al.*, 2002; Miller *et al.*, 1980).

The review revealed that the BOD, COD, and TSS of the paper mill exceed the permissible limit (Figure 5). Because of its alarming level of BOD, COD, TSS, and other pollution parameters of paper mill effluent are very toxic to phytoplankton and zooplankton. It also induces the growth of fecal coliform bacteria that is hazardous for the living organism (Gauthier and Archibald, 2001; Chandra *et al.*, 2006).

Pulp and paper mill wastewater contains a lot of complex organic pollutants named cellulose, hemicellulose, lignin, resins, fatty acids along with other phenolic compounds (Lacorte *et al.*, 2003). These organic toxic pollutants cause ecosystem degradation, different diseases of the aquatic organism as well as a health hazard in the human body (Table 4). Many studies reveal that wastewater discharged from pulp and paper mill has a toxic/lethal impact on the fishes (Johnsen *et al.*, 1998; Kovacs *et al.*, 2002; Yen *et al.*, 1996). It is indicated that lignin derivatives exhibit toxic potential (Pessala *et al.*, 2004). Chlorine dioxide, chlorine, hydrogen peroxide, ozone, etc. are used singly or combined for bleaching of pulp, where phenols, lignin, resin, acids, etc. get chlorinated and convert into very harmful xenobiotics.

The chemicals including chlorinated phenols, monomeric phenols, enol ethers, acetaldehyde, formic acid, acetic acid, mercaptides, quinone derivatives, methanol, methylglyoxal, furfural, etc. are usually present in the pulp and paper mills' wastewater. These are the degrading compounds of cellulose, hemicellulose lignin, and wood extractives. In the pulp and paper mills, almost 300 organochlorine compounds were identified, and another hundred were not identified (Singh et al. 2019). Labunska et al. (2001) showed that pulp and paper mills effluent contained organochlorine compounds that harm the ecology of the water environment.

Table 3: Physicochemical parameters of paper mill effluent

Parameters	Reference						Permissible limit: USEPA, 2002
	Devi *et al.*, 2011	Singh and Chandra, 2019	Giri *et al.*, 2014	Ahirwar & Gupta, 2017	Kumar *et al.*, 2003	Chandra *et al.*, 2018	
Color (Co-Pt)	-	625	-	Milky colloidal	Dark brown	2500	-
pH	6.45	7.0	7.32	6.2	7.8	-	5-9
Turbidity	-	-	-	394.5	-	-	-
EC	2650	-	1290	1941.8	1120	-	-
BOD	203.75	2700	306	-	1638	6000	40
COD	1145.5	3000	4357	-	3873	17999	120
TDS	996.5	110	1380	1027.5	2565	560	-
TSS	86	-	-	165.5	-	56	35
TS	-	-	1840	-	4870	616	-
Alkalinity	302.35	-	-	123.8	-	-	-
Hardness	-	-	-	586.3	-	-	-
HCO_3^-	-	-	440	-	-	-	-
NH_4- N	-	-	7.92	-	-	-	-
Total-N	-	103	-	-	-	143	143
Cl^-	45.175	1.23	347.6	-	-	2.04	1500
PO_4^{3-}	0.0	-	0.98	-	-	-	-
NO_3^-	-	-	2.48	-	-	-	-
SO_4^{2-}	8.5	1280	0.27	-	-	1692	250
Na^+	-	25	826	106.8	19.6	64	200
K^+	-	1.38	334	18.5	47.3	7.8	200
Ca^{2+}	-	-	2346	190.5	10.9	-	-
Mg^{2+}	-	-	51	30.5	8.2	-	-
Zn	0.2088	0.27	0.04	-	-	13.90	2.0
Mn	-	0.07	0.25	-	-	11	0.2
Pb	0.0075	1.05	0.081	-	-	-	0.05
Co	-	--	0.021	-	-	-	-
Ni	0.0532	0.19	0.019	-	-	3.3	0.1
Cu	0.076	0.09	0.006	-	-	2.15	0.5
Fe	0.1113	1.05	0.73	-	-	67.53	2.0
Cr	-	0.11	0.007	-	-	2.30	0.05
Cd	0.0965	0.02	0.004	-	-	0.255	0.01
Lignin	-	1550	-	-	1235.7	46000	0.05
Chlorophenol	-	195	-	-	-	203	3.0
Total phenol	-	389	-	-	-	413	0.5

Due to the presence of chlorine in phenol rings, chlorophenols are well-known toxicants that were produced in the pulping and bleaching process of the paper mills (Michalowicz and Duda, 2007). The reaction of chlorine and the breaking of lignin, phenolic, and the other alkaloids in the pulping process produced the toxicants (Knuutinen, 1984). Estrogenic and mutagenic toxicity of chlorophenol were reported (Michalowicz and Duda, 2007). A report showed that chlorophenols produced dioxins and furans during incineration and metabolism. These chemicals are toxic and develop cancer, damage the immune system, and interfere with hormone functions (Criado *et al.*, 2004).

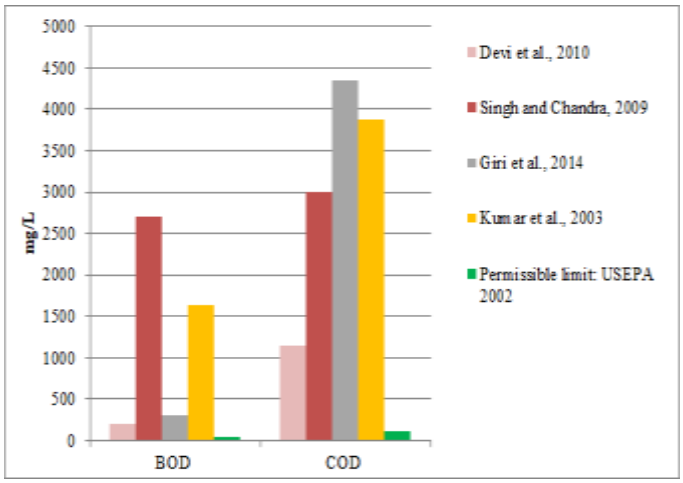

Figure 5: Paper mill wastewater BOD and COD comparison with USEPA 2002 permissible limit.

EFFECT ON WATER, SOIL, CROPS AND LIVESTOCK

Several fish diseases, including sexual immaturity, reduce reproduction, depression in sexual characteristics, and smaller gonads were reported due to the discharge of paper industries effluent into water bodies. Besides, there were several compliances associated with the growth of sewage fungus on river and canals, photosynthetic activity in aquatic plants, etc. were effects of discharging the sugar, pulp and paper mills effluent on surface water body (Rahim and Mostafa, 2021; Munkittrick et al. 1997; Webb. 1985; Singh et al. 2004; Ruggiero et al. 1989). Nutrient imbalance in crops, deterioration in soil structure, increasing soil salinity, and finally lower crop production have been reported due to sodium, calcium, magnesium, chloride, and sulfate ions which are found excessive in pulp and paper mill wastewater (Sundari and Kanakarni. 2001). Pulp and paper mills effluent contain a high volume of BOD, COD, cation, anion, toxic organic compounds, and heavy metals. These pollutants accumulate in soil and deteriorate the soil quality. Pulp mills effluent causes increasing pH of the soil, changing soil color & texture, imbalance of macro and micronutrients in soil, depletion of oxygen supply in the soil, adverse effect on seedling growth, decrease in germination percentage, etc. (Singh et al. 2019)

Hampering seed germination of paddy crop, maize, and sunflower; growth, quality, and yield degradation of the crop; bioaccumulation of toxic pollutants into farm animals and livestock through the food chain, etc. have been reported because of the pulp and paper mill effluent (Somashekar *et al.*, 1984; Rajamani and Oblisami, 1979; Sahai *et al.*, 1985; Singh *et al.*, 2002; Dutta and Boissya, 1997; Mishra and Sahoo, 1989; Singh *et al.*, 2019).

CHALLENGES OF EFFLUENT MANAGEMENT

There are many challenges for sustainable development in this sector and maintaining environmentally friendly industrial management. The owner of the industries is very concerned about the profit but similarly unaware of environmental degradation. Moreover, people lived surrounding the industry are also not much aware of the pollutant released and their fate and effects on fish, animals, and humans as well.

Pollution control and regulatory authorities have a lack of skilled manpower, monitoring, and measuring instruments and equipment. Finally, the available fund for pollution and control management is another major problem. Despite several challenges of effluent management, it is imperative to install an effective and efficient effluent treatment plant (ETP) for every industry, and the owner and authority should come forward with an environmentally friendly approach for sustainable paper industries.

Table 4: Toxic organic compounds of pulp and paper industry effluent

Organic compounds	Affected species	Toxicity	Pollution Source	Reference
Organochlorine compounds (AOX)	Aquatic organism	carcinogenicity and mutagenicity, acute toxicity	chlorine bleaching wastewater	Deshmukh *et al.*, 2009; Thompson *et al.*, 2001
Hydroxy furanones with mono-, di-, and trichloromethyl groups	Ames test	mutagenicity	chlorine bleaching	Franzén and Kronberg, 1994
Resin acids , biocides such as methylene bis thiocyanate (MBT) and 2-(thiocyanomethylthio)benzothiazole (TCMTB)	algae, fish, and daphnia	acutely toxic	Kraft pulping effluent	Kruzynski and Birtwell, 1994; Reemtsma *et al.*, 1995
Nonylphenol, bisphenol A (BPA), estrone, estradiol, triclosan (TCS), PCBs, PAHs, PCDD/PCDF	Fish	Growth inhibition	Wastewater	Fang *et al.*, 2012
Methyl dehydroabietate, ethyl abietate, dehydroabietic acid, abietic acid	*Saccharomyces cerevisiae*	Antiestrogenic activity	Wastewater	Terasaki *et al.*, 2009
Nitrilotriacetic acid, EDTA, DTPA	Human	Hydronephrosis and nephromegaly	Wastewater	Lee *et al.*, 1996
Chlorinated dibenzo-p dioxins and chlorinated dibenzofurans	Aquatic organism	mutagenic tendencies, damage to genetic structures	wastewater	Oanh *et al.*, 1999; Ali and Sreekrishnan, 2001
Dimethyl Diphenyl Methane, dimethylphenyl ethane, di-iso-propyl naphthalene, benzyloxy benzene	Fish	Fish tissue, Reproductive toxicity	Wastewater	Terasaki *et al.*, 2012
Chlorophenols, chlor guaiacol s, chlorocatechols	Fish	fish bile, Carcinogen	Bleaching Effluent	Soderstrom *et al.*, 1994
Resin acids (RAs), unsaturated fatty acid	Fish and aquatic organism	Genotoxicity, Bioaccumulation	Pulping Effluent	Lacorte *et al.*, 2003
2,4,7,9-Tetramethyl-5-decyne-4,7-diol (TMDD)	Aquatic organism	Ecotoxic	Paper recycling wastewater	Guedez and Püttmann, 2013

CONCLUSIONS

The pulp and paper industry is one of the most growing-up industries in the world. Many developing countries are boosting up their paper production rapidly to mitigate the present need. This sector uses a large volume of water. Paper mills discharge untreated or partially treated effluents into the surface water bodies like rivers, canals, and estuaries. The worldwide pulp and paper mills industry yearly discharged effluents was approximately 69094 million m^3. It contained higher values of BOD, COD, EC, TSS, toxic organic compounds, and heavy metals. It caused harm to aquatic life and human healths. There must be an effluent treatment plant installed in each and every pulp and paper industry considering modern technological facilities including ultra-violate (UV) radiation technology, membrane technology combined with biological treatment to ensure safe surface water quality around the industrial areas and thus, help to develop an environmentally friendly ecosystem. Moreover, appropriate and modern rules, regulations, and acts should be formulated, along with publicity and awareness programs would be taken. Further, pollution monitoring and controlling authorities should have a closer look to ensure sustainable pulp and paper production.

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