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A Comparative Study on Physico-Chemical Properties of Pulp and Paper Mill Effluent

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Abstract
In the present study physico-chemical properties of small and large scale pulp and paper industry effluents are compared in relation to their hazardous impact on environment. Effluent samples were collected from different sections of bleaching unit of small and large scale pulp and paper mills and then analyzed for various physicochemical parameters such as pH, colour, biochemical oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids (TSS), total dissolved solids (TDS), and turbidity. Highly significant differences were thus found at all levels of both the pulp and paper mills. The results indicate that pulp and paper industrial effluents are highly polluted and had variation in the pollution load on different raw material used. Agro based small scale pulp and paper mill is way more contributor of pollution than wood based large scale.
Keywords: BOD, COD, Paper mill effluent, TDS

I. Introduction

An enormous industrial growth has taken place throughout the world in the past few decades, to fulfill the increased demand of human civilization, which has created an overexploitation of available resources and caused pollution of water, land, and air. Pulp and paper industry is a highly energy and water intensive industry and also one of the biggest polluting sectors of water bodies (being the sixth largest water polluting sector as per a previous study by Pokhrel and Viraraghavan [1]. During paper production, large amount of waste water is generated that requires safe disposal. Typically in India, approximately 75% of the total fresh water supplied to pulp and paper industries emerges as waste water. In comparison to other industries, fresh water requirement in pulp and paper industry is quiet high, i.e, 150-200 m^3 per ton of paper produced [2].

Pulp and paper industry is mainly categorized into three types according to their production capacity and raw materials usage i.e. small scale, medium scale and large scale. The small scale paper mills set up in the early seventies almost exclusively use agro waste/residues as raw materials for paper production whereas large scale mills, so far, have mainly been based on forest based woody raw material for paper production. As agro waste/residues such as rice straw, wheat straw and bagasse are relatively short cycled, regenerative and abundant, on the other hand the availability of forest based raw material is rather limited. Small scale paper mills suffer from high production costs, uneconomic operation, low quality and negative impacts on the environment. Furthermore, most small scale pulp and paper mills cannot economically provide chemical recovery and pollution control systems.

Pulp and paper manufacturing process generates highly polluting waste water/effluent during various processing stages such as raw material preparation, pulping, pulp washing, screening, bleaching and, coating operation etc. Pulp bleaching generates toxic substances as it utilize oxygen, hydrogen peroxide, ozone, sodium hypochlorite, chlorine dioxide, chlorine, and other chemicals for whitening and bighting the pulp. Bleaching processes also release chlorinated lignosulphonic acids, chlorinated resin acids, chlorinated phenols and chlorinated hydrocarbons in the pulp and paper mill effluent [1, 3, 4, 5, and 6].

The characteristic problems arise due to discharge of pulp and paper mill effluents to water bodies are alarming, such as pH change, colour of water bodies, increased levels of biochemical oxygen demand (BOD) and chemical oxygen demand (COD), also being contaminated with suspended solids (SS), fatty acids, tannins, resin acids, sulphur and sulphur compounds, adsorbable organic halid (AOX) etc. The pollutants discharged from the pulp and paper industry affect all aspects of the environment. Presence of toxic pollutants poses serious threat to flora and fauna of aquatic system [7 and 8]. Furthermore some compounds in the effluents are resistant to biodegradation and can bioaccumulate in the aquatic food chain [9]. The paper mill effluents also have drastic impact on human health leading to outbreak of diseases such as diarrhoea, vomiting, headache, nausea and eye irritation [10].

Pulp and paper mills effluent is complex in nature and its characteristics varies from mill to mill depending on numerous factor including raw materials finish and process, paper manufacturing technology, washing, cooking, bleaching etc. Due to the severe toxic effects of pulp and paper mill effluent there is an urgent need to reduce pollution load before it discharged into the environment. Considering the need to characterize the physicochemical properties of pulp and paper mill effluent, the present study was undertaken with the objective to portray the physico-chemical parameters of two pulp and paper mill effluents; one is small scale and other large scale. For efficient treatment technologies for the same, it was important to assess the actual load of pollution, in terms of selected parameters, exerted by each individual processing unit of mills differing in raw material utilization as well as scale of operation [1]. The present study shows an overview of the characteristics of wastewater emanating from different processing units of two different types of Indian pulp and paper mills. The data would be useful in developing innovative effluent treatment techniques for pulp and paper industries.

II. Materials and Methods

2.1 Sample collection

For the present study, samples were collected from various processing units of both the small and large scale pulp and paper mills. The sampling sites were bleaching unit (chlorination stage, extraction stage, combined bleach plant effluent) and combined plant effluent (effluent generated from all the processes involved in paper manufacturing). Samples were collected in clean containers and transported to the laboratory within 5-6 h and stored below 4°C till use. Samples were collected over the year in three seasons namely summer (March-June), monsoon (July-October) and winter (November–February).

2.2 Mill description

The study was conducted on the effluents collected from two pulp and paper mills situated in Uttarakhand (Mill A) and Uttar Pradesh (Mill B), India.

2.2.1 Mill A

Mill A is a small scale agro based paper mill which produces writing and printing papers and has a production capacity of 30 tons per day. This mill uses soda process for pulping by using sugarcane baggase and wheat straw as raw material. Chemical charge during pulping process is 12% for wheat straw and 16% for bagasse. Unbleached pulp yield is about 50 % having kappa number 20-22. Bleaching is performed by conventional CEHH bleaching sequence. Bleached plant effluent generation is 50-60m^3 /ton and combined mill effluent is 100-125m^3 /ton paper production.

2.2.2 Mill B

This is an old and large scale basically wood based paper mill, and has an installed capacity of 200 tpd. It produces printing, chart absorbent and kraft paper. The mill uses kraft pulping process by using raw material such as hardwood like poplar, bamboo and eucalyptus and small quantities of waste paper and wood pulp for its production. Oxygen bleaching is performed followed by chemical bleaching which is carried out in four stages, viz., CEHH. Each step is preceded and followed by counter current washing with fresh water. A total of 25,000 m^3 of water is consumed for the entire process of pulping and papermaking whereas around 23,000 m^3 is discharged as wastewater.

2.3 Chemical

All the chemicals were of analytical grade and procured from M/s. s.d. fine. Chem. ltd. All the solutions were prepared in double distilled water and were preserved in Schott Duran bottle. The laboratory glass used, were washed with detergents and rinsed with distilled water and then oven baked at 200°C overnight, prior to use.

2.4 Characterization of effluent

The effluent collected from both the mill i.e Agro based (paper mill A) and wood based (paper mill B) were characterized for pH, colour, COD, TDS, BOD, suspended solids, turbidity as per the American public Health Association (APHA) standard methods [11]. The experiments were carried out in triplicates in order to access the repeatability of the results. pH of the effluent samples were measured by using Microprocessor pH meter (Labtronics). Colour of the samples was analyzed by spectrophotometric method. Turbidity of the effluent was measured by turbidity tube.

III. Results and discussion

3.1 pH

Table 1 and 3 summarize the variation among the pH of the samples collected from different sites of mill A and mill B (chlorination, extraction, combined bleach plant effluent, combined plant effluent) in different season i.e summer, winter and monsoon. pH showed high variation according to process used in bleaching. pH of the sample vary from site to site due to the utilization of different chemicals in each process. The mean value of pH for site C, E, CBP, CPE was 2.6, 9.1, 7.3 and 8.2 respectively in mill A and 2.7, 10.5, 6.4, and 9.4 respectively in mill B. The mean pH value varied from 2.6 to 10.5 in all the samples collected from four different stages. The minimum mean value of pH (during summer, winter and monsoon season) was found 2.6 in the effluent sample collected from chlorination section of mill A which would be due to the reaction of chlorine with water. Chlorine reacts with water and forms hypochlorous acid and hydrochloric acid. Hypochlorous acid may break down into a hydrogen ion and a hypochlorite ion, which lowers the pH of the effluent. A maximum mean pH value 10.5 was obtained in extraction stage of mill B. This would be due to the addition of alkali for the precipitation of total solids. The alkali reacts with the water and produces higher no. of hydroxyl ions which raises the pH of effluent and makes it alkaline in nature. The data from table 1 and table 2 reveals that the mean pH of chlorination stage in both the mills is almost same where as the mean pH of E, CBP, CPE stage differs from each other which would be due to utilization of different chemicals.

3.2 Colour

The colour is usually the first contaminant to be recognized in wastewater/effluent that affects the aesthetics, water transparency and gas solubility of water bodies [12]. The colour of the effluent mainly arises due to the presence of low and high molecular weight chlorinated organic compound generated from the lignin degradation product which are produced during different stages like pulping, bleaching and alkali extraction. Table 1 and 3 shows the variation in colour in different sections. Visibly, the effluent had a dark brown to yellowish brown appearance at the time of collection of samples. The mean value of colour for site C, E, CBP, and CPE was 457.3, 2175.4, 898.3 and 4752.6 PCU (platinum cobalt unit) respectively in mill A and 794.7, 1709.2, 1125 and 2531 PCU respectively in mill B. The data presented in table 1 revealed that CPE has maximum colour value followed by E, CBP and C stage. This much high colour of combined plant effluent of mill A may be due to presence of some amount of weak black liquor.

3.3 Chemical Oxygen Demand (COD)

Chemical oxygen demand measures amount of oxygen required to breakdown both organic and inorganic matters. The mean COD value of the samples from site C, E, CBP, and CPE were recorded as 835.6, 1143.3, 824.6 and 2664.2 mg/l respectively in case of mill A and 930, 1132.5, 802.1 and 1566.7 mg/l in case of mill B. Combined plant effluent of mill A had maximum COD value which may be again due to presence of weak black liquor in the stream. High COD levels indicate toxic state of the effluent along with the presence of recalcitrant organic substances [13]. COD of mill A effluent was high except chlorination stage, which could be due to the different raw materials and processing chemicals were used during pulping (Table 1 and 3).

3.4 Biological Oxygen Demand (BOD)

Biological oxygen demand measures amount of oxygen required by microorganisms for breaking down organic matter. BOD is the most commonly used parameter for determining the pollution load municipal or industrial discharge. BOD can also be used to evaluate the efficiency of treatment processes, and is an indirect measure of biodegradable organic compounds in water. The mean BOD value for site C, E, CBP, and CPE was 232.7, 183.5, 339.2 and 670.8 mg/l respectively in case of mill A whereas it was 340.8, 456.3, 306.3 and 600.8 mg/l respectively in case of mill B (Table 1 and 3). BOD has the maximum mean value in CPE stage of mill A and minimum in the E stage for the same mill A. BOD of the effluents, were found to be much higher in all the sections than the permissible limits for inland surface water which is 30 mg/l whereas it is 100mg/l for land irrigation according to regulatory agency [14]. The higher mean value BOD is mainly due to the presence of organic matter present in the effluent which depends upon the rare material used for paper making.

3.5 Total dissolved solids (TDS)

Total dissolved solids are measurement of inorganic salts, organic matter and other dissolved materials in water [7]. In general, TDS is the sum of the cations and anions in water. Ions and ionic compounds making up TDS usually include carbonate, bicarbonate, chloride, fluoride, sulphate, phosphate, nitrate, calcium, magnesium, sodium, and potassium**.** The values obtained for TDS were more than WHO (World Health Organisation) standard of 2000mg/l for the discharge of wastewater into surface water. The data reveals that the extraction stage of mill B is highly polluted with respect to TDS content among all the stages which could be due to solubilisation of maximum chlorolignin compounds present in the E stage of mill B. Various disorders like dysfunction of elementary canal, respiratory system, nervous system, coronary system besides, causing miscarriage and cancer have been reported by consumption of water with high concentrations of total dissolved solids has been reported [15]. Various negative impacts of high TDS have been seen in fish species, especially at fertilization [16]. The United States Environmental Protection Agency recommends treatment when TDS concentrations exceed 500 mg/l or 500 ppm (parts per million) [17].

3.6 Total suspended solids (TSS)

TSS concentration of all the sections of both the mills was summarized in table-2 and 4. Suspended solids are present in sanitary wastewater and many types of industrial wastewater. As levels of TSS increase, a water body begins to lose its ability to support a diversity of aquatic life. Suspended solids absorb heat from sunlight, which increases water temperature and subsequently decreases levels of dissolved oxygen in the warmer water holds less oxygen than the cold water. The mean value of TSS of the effluent from site C, E, CBP, and CPE was 399.2, 367.5, 389.2 and 1122.5 mg/l respectively in case of mill A whereas it was 409.2, 810.8, 222.5 and 301.7 mg/l respectively in case of mill B. Data reveals that mill A has maximum TSS content in CPE stage which would be due to inclusion of effluent from all the other sections.

3.7 Turbidity

Turbidity is an expression of the optical property that causes light to be scattered rather than transmitted in straight lines through the sample. In water, turbidity is caused by suspended matter, such as organic and inorganic matter, soluble colored organic compounds, and plankton. Turbidity is a measure of water clarity which shows how much the material suspended in water decreases the passage of light through the water. Turbidity can affect the colour of the water. Higher turbidity also reduces the amount of light penetrating the water, which reduces photosynthesis and the production of dissolved oxygen. The mean value of turbidity of the effluent from site C, E, CBP, and CPE was 148.6, 155.8, 146.7 and 494.2 NTU respectively in case of mill A and 76.6, 97.8, 106.3 and 351.7 NTU (Nephlometric Turbidity Unit) respectively in case of mill B. The data presents in table 2 and 4 reveals that the mean value of turbidity is higher for all the stages for mill B in comparison to all the stages of mill A. The higher mean value of turbidity for mill B is mainly due to the addition of different chemicals during pulping and bleaching processes.

IV. Conclusion

Pulp and paper industry effluents are highly polluted industries in India. Small and large scale pulp and paper mills which have different production capacity as well as different raw materials, adopt different processes that lead to radical differences in the physico-chemical properties of effluents. Such polluted effluents must be treated properly before being discharged into the drainage channel, to minimize the effect of various pollutants on the environment. On the basis of results reported herein it can be concluded that the effluent discharged from both the paper industries is highly polluted and has exceeding values as prescribed by the standards of regulatory agency of India. It is further stated that the pollutants generated during different stages from paper industries can be minimized either by replacing some existing pulping and bleaching techniques like bio-pulping, bio-bleaching, TCF (Total Chlorine Free bleaching), ECF (Elemental Chlorine Free bleaching) and ozone bleaching or by treatment of the effluent by physico-chemical or biological methods. The data bank generated herein by this monitoring study of pulp and paper mill effluent are collected from Agro-based and one from wood based could successfully be used in prediction of their toxicity and effective management. Hence proper strategies should be designed to counter treat the waste load borne by effluents prior to their disposal into the environment.

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Table 1. pH, Colour, COD and BOD of effluent emanating from different sections of small scale paper mill (mill A)

Sample		pH				Colour (PCU)				COD (mg/l)				BOD (mg/l)			
Season	***S.N.***	***C***	***E***	***CBP***	***CPE***	***C***	***E***	***CBP***	***CPE***	***C***	***E***	***CBP***	***CPE***	***C***	***E***	***CBP***	***CPE***
Summer	1	3.05	9.97	7.8	8.5	488	2234	920	4943	880	1160	840	2820	200	184	360	750
	2	2.8	9	7.5	8.3	450	2200	890	4500	750	1100	820	2690	192	181	290	720
	3	2.6	8.5	7.4	8.5	520	2243	934	5000	920	1120	835	2600	250	187	320	630
	4	2.5	7.9	7.3	8.1	490	2238	965	4578	800	1090	810	2700	225	200	350	710
Winter	5	2.7	9.5	7.5	8.3	520	2300	970	5010	920	1210	860	2790	280	200	400	750
	6	2.5	9.3	7.3	8.1	500	2280	920	4920	880	1190	840	2650	250	180	390	650
	7	2.7	9.1	7.3	8.3	480	2260	980	4780	780	1200	850	2710	290	210	340	640
	8	2.2	9.5	6.9	8	510	2320	990	5000	850	1230	820	2700	300	220	360	700
Monsoon	9	2.5	9.3	7.3	8.1	370	2100	850	4500	810	1100	810	2400	190	160	320	830
	10	2.4	9.4	7.1	8	350	1800	780	4400	800	1090	790	2590	195	150	290	520
	11	2.3	9.1	7.2	8	420	2200	730	5000	830	1110	800	2700	200	170	310	630
	12	2.5	9.2	7.1	8.1	390	1930	850	4400	800	1120	820	2620	220	160	340	520
Mean		**2.6**	**9.1**	**7.3**	**8.2**	**457.3**	**2175.4**	**898.3**	**4752.6**	**835.0**	**1143.3**	**824.6**	**2664.2**	**232.7**	**183.5**	**339.2**	**670.8**
SD		**±0.2**	**±0.5**	**±0.2**	**±0.2**	**±57.8**	**±151.2**	**±78.5**	**±245.2**	**±52.7**	**±49.4**	**±19.9**	**±103.4**	**±38.6**	**±20.5**	**±33.8**	**±88.0**

SN *- Sample Number,* ***C****- Chlorination Stage,* ***E*** *- Extraction Stage,* ***CBP*** *- Combined Bleach Plant,* ***CPE*** *- Combined Plant Effluent,* ***SD*** *– Standard Deviation*

Table 2. TDS, TSS and Turbidity of effluent emanating from different sections of small scale paper mill (mill A)

Sample		TDS (mg/l)				TSS (mg/l)				Turbidity (NTU)			
Season	***SN***	***C***	***E***	***CBP***	***CPE***	***C***	***E***	***CBP***	***CPE***	***C***	***E***	***CBP***	***CPE***
Summer	1	3800	4000	2200	2600	400	400	400	1200	128	150	125	480
	2	3600	3800	2100	2590	350	350	415	1140	140	165	140	500
	3	3850	4020	2250	2610	460	420	415	1220	125	140	130	430
	4	3890	4050	2300	2650	500	450	430	1210	150	150	150	450
Winter	5	3850	4100	2280	2765	460	410	450	1210	150	180	170	550

	6	3750	4090	2100	2700	440	380	420	1200	140	155	160	450
	7	3800	4200	2300	2721	460	420	440	1220	165	165	155	480
	8	3880	4350	2380	2740	510	440	460	1210	170	175	140	500
Monsoon	9	3675	3850	2160	2520	300	300	300	1100	140	170	150	600
	10	3600	3860	2170	2430	250	280	290	1050	170	140	155	450
	11	3680	3890	2232	2550	260	250	320	1240	155	130	155	540
	12	3692	3910	2246	2580	400	310	330	1190	150	150	130	500
Mean		**3755.6**	**4010**	**2226.5**	**2621.3**	**399.2**	**367.5**	**389.2**	**1182.5**	**148.6**	**155.8**	**146.7**	**494.2**
SD		**±100.0**	**±154.0**	**±80.9**	**±94.4**	**±86.2**	**±64.7**	**±58.8**	**±54.2**	**±14.2**	**±14.7**	**±13.1**	**±47.3**

SN *- Sample Number,* ***C****- Chlorination Stage,* ***E*** *- Extraction Stage,* ***CBP*** *- Combined Bleach Plant,* ***CPE*** *- Combined Plant Effluent,* ***SD*** *– Standard Deviation*

Table 3. pH, Colour, COD and BOD of effluent emanating from different sections of large scale paper mill (mill B)

Sample		**pH**				**Colour (PCU)**				**COD (mg/l)**				**BOD (mg/l)**			
Season	***SN***	***C***	***E***	***CBP***	***CPE***	***C***	***E***	***CBP***	***CPE***	***C***	***E***	***CBP***	***CPE***	***C***	***E***	***CBP***	***CPE***
Summer	1	3.05	10.5	6.5	9.5	800	1700	1200	2500	930	1120	800	1580	350	430	300	600
	2	2.6	10.1	6.4	9.1	780	1650	1140	2400	910	1100	780	1820	310	410	250	540
	3	2.5	10.3	6.5	9.3	830	1600	1220	2450	950	1130	810	1390	380	440	310	610
	4	2.6	10.5	6.4	9.3	800	1750	1300	2530	900	1150	830	1650	320	460	320	640
Winter	5	2.8	10.5	6.5	9.5	820	1850	1270	2650	950	1150	850	1580	380	480	320	650
	6	2.8	11	6.5	9.5	800	1810	1200	2630	930	1110	830	1620	350	450	300	630
	7	2.7	10.4	6.3	9.4	830	1860	1280	2670	960	1160	860	2000	390	510	340	660
	8	2.6	10.5	6.3	9.5	860	1890	1290	2695	990	1210	880	1790	420	530	380	680
Monsoon	9	2.8	10.5	6.5	9.5	750	1590	1140	2460	900	1100	740	1460	280	410	280	540
	10	2.8	10.5	6.4	9.5	710	1540	110	2410	890	1090	710	1110	260	430	265	520
	11	2.7	10.4	6.4	9.4	760	1620	1160	2485	910	1120	750	1340	310	455	290	560
	12	2.8	10.5	6.5	9.5	795	1650	1195	2492	940	1150	785	1460	340	470	320	580

Mean	**2.7**	**10.5**	**6.4**	**9.4**	**794.6**	**1709.2**	**1125.4**	**2531**	**930**	**1132.5**	**802.1**	**1566.7**	**340.8**	**456.3**	**306.3**	**600.8**
SD	**±0.14**	**±0.20**	**±0.07**	**±0.12**	**±38.81**	**±114.34**	**±310.91**	**±99.24**	**±28.28**	**±32.18**	**±49.31**	**±228.38**	**±45.18**	**±35.42**	**±32.99**	**±50.57**

SN *- Sample Number,* ***C****- Chlorination Stage,* ***E*** *- Extraction Stage,* ***CBP*** *- Combined Bleach Plant,* ***CPE*** *- Combined Plant Effluent,* ***SD*** *– Standard Deviation*

Table 4. TDS, TSS and Turbidity of effluent emanating from different sections of large scale paper mill (mill B)

Sample		**TDS (mg/l)**				**TSS (mg/l)**				**Turbidity (NTU)**			
Season	***SN***	***C***	***E***	***CBP***	***CPE***	***C***	***E***	***CBP***	***CPE***	***C***	***E***	***CBP***	***CPE***
Summer	1	2400	2000	1600	2400	400	800	200	300	75	94	107	350
	2	2350	2220	1630	2450	380	750	180	180	73	90	95	310
	3	2390	2400	1700	2300	420	810	210	310	79	98	102	370
	4	2400	2000	1680	2420	450	830	240	350	80	100	110	410
Winter	5	2400	2100	1750	2490	400	850	250	320	80	100	110	365
	6	2310	2130	1700	2480	390	810	235	310	78	95	100	355
	7	2360	2200	1730	2420	410	800	250	340	86	99	118	370
	8	2300	2100	1760	2490	400	880	280	365	90	110	121	375
Monsoon	9	2200	1900	1500	2300	400	790	195	280	68	90	100	320
	10	2140	2120	1480	2430	390	760	190	260	65	91	101	315
	11	2180	2200	1600	2220	420	830	210	295	71	101	101	330
	12	2200	1910	1520	2290	450	820	230	310	74	105	110	350
Mean		**2302.5**	**2106.7**	**1637.5**	**2390.8**	**409.2**	**810.8**	**222.5**	**301.7**	**76.6**	**97.8**	**106.3**	**351.7**
SD		**±93.2**	**±135.8**	**±94.1**	**±86.7**	**±21.4**	**±34.3**	**±28.5**	**±46.0**	**±6.8**	**±5.9**	**±7.5**	**±27.9**

SN *- Sample Number,* ***C****- Chlorination Stage,* ***E*** *- Extraction Stage,* ***CBP*** *- Combined Bleach Plant,* ***CPE*** *- Combined Plant Effluent,* ***SD*** *– Standard Deviation*