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Seasonal variations in different physico-chemical parameters of the effluents of Century Pulp and Paper Mill, Lal Kuan, Uttarakhand

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Abstract: *The present study was undertaken with the objective to study the characteristics of the effluent of Century Pulp and Paper Mill, Lalkuan (Uttarakhand) in different seasons. The variations in the physicochemical characteristics were observed and monitored upto 12 months at three different sites. Mean values of temperature, pH, chlorides and total phenols of the effluent were found below, whereas colour,* BOD_5*, COD and lignin concentrations were above the minimum national standards (MINAS).*

Key words: *Pulp and paper industry, Effluent, Physicochemical parameters, Seasonal variation, MINAS*

Introduction

India ranks 20th among the paper producing countries of the world (Mahajan, 1985). There are more than 150 paper and board mills in the country with an installed capacity of nearly three million tonnes yr^{-1}, of which 36 are the large mills with a production capacity of >55 tonnes d^{-1}, and the rest are small mills with production capacity of <30 tonnes d^{-1} (Sastri, 1986; Subramanyam, 1990).

Huge quantities of fresh water was required for the production of paper. It has been estimated that as much as 250-450 m^3 of water is required for production of one tonne of paper (Mahajan, 1985). The volume and characteristics of pulp and paper mill waste water (effluents) depend upon the type of manufacturing process adopted and the extent of reuse of water employed in the plant. The large pulp and paper mills equipped with soda recovery, discharge about 270 to 450 liters effluent kg^{-1} paper, containing 40 to 50 g lignin kg^{-1} bleached paper produced. Contrary to this, the small paper mills without soda recovery, discharge nearly 300 to 400 liters of black liquor effluent containing 200 to 250 g lignin kg^{-1} of paper manufactured (Garg and Modi, 1999).

During cellulose pulp production, lignin is removed in the cooking stage and in subsequent bleaching stage. In the conventional bleaching, chlorolignins as well as low molecular weight chlorinated aromatic and aliphatic compounds are formed. Most of the chlorinated compounds are produced in the first chlorination stage, and in the subsequent alkaline extraction stage of the bleaching sequence (Osterberg and Lindstrom, 1985). Many of the low molecular weight compounds, especially chlorinated guaiacols and phenols are toxic to the aquatic flora and fauna.

The pulp and paper industries every year generate more than 700 billion gallons of highly coloured and toxic effluents mainly containing high molecular weight modified and chlorinated lignin. About 300 different chlorinated organic compounds in bleached pulp mill effluent have been identified till date. About 200 of these compounds are chlorinated resin acids, phenols and dioxins (Huynh *et al.*, 1985).

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 physicochemical parameters of Century Pulp and Paper Mill effluents for one year duration at different sites.

Materials and Methods

The study was conducted on the combined effluents released from the Century Pulp and Paper Mill, Lalkuan, Nainital (Uttarakhand) situated at about 7 km from G.B. Pant University of Agriculture and Technology, Pantnagar on the Bareilly Nainital highway. This industry was established in June 1984 and is engaged in the manufacture of some finest varieties of writing and printing paper using kraft process. The mill has three drains, which combine to form one main effluent channel (combined effluent). Average discharge of effluent from the mill is 7000 gallons per tonne of paper produced. The huge amount of this highly coloured effluent moves through the villages, agricultural lands and nearby forests and ultimately joins the river Gola, 15 km southeast of the mill.

For the study of seasonal variations on physicochemical characteristics of paper mill effluent three sites S_1, S_2 and S_3 were selected along the effluent channel released from the Century Pulp and Paper Mill, Lalkuan for a 12 months period.

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These sites were situated from 1.0, 6.0 and 7.0 km distance from the origin of effluent. The sites were selected on the basis of extent of dilution. Samples were collected from the sampling sites between 10 a.m. to 2 p.m. in sterilized plastic carboy, brought to the laboratory and analysed for measurement of various physicochemical parameters.

Temperature of the effluents and ambient temperature were measured using portable digital multi-stem thermometer (Hanna Instrument Co., Italy) with external sensing probe. pH of the effluent samples were measured using digital portable pH meter (Hanna Instrument Co., Italy). The pH meter was allowed to stabilize before making the final reading. The electrical conductivity (EC) of effluent water was measured on the sampling site using a digital portable conductivity meter at 25°C (Hanna Instrument Co., Italy). Colour units were measured spectrophotometrically using the method of Bajpai *et al.* (1993).

Lignin content in the effluent water was measured by the modified Nitrosation method (Pearl and Benson, 1940). The absorbance was read at 430 nm using a spectrophotometer. Total phenols in the effluent sample were measured by using 4-aminoantipyrine colorimetric method (APHA, 1995). Biochemical oxygen demand (BOD) and chemical oxygen demand (COD) of the effluent sample were measured according to APHA (1995). The chloride content of the effluent sample was measured by argentometric method (APHA, 1995).

Results and Discussion

Monthly variations among the physical parameters of Century Pulp and Paper Mill effluents are summarized in Table 1. The mean value of atmospheric temperature for site S_1, S_2 and S_3 was 29.8, 27.7 and 28.6°C, respectively. Atmospheric temperature was maximum (41.6°C) in the month of June and minimum (14.8°C) in the month of December. The mean value of effluent temperature for site S_1, S_2 and S_3 was 33.0, 28.8 and 27.3°C, respectively. Maximum effluent temperature (35.8°C) was recorded in the month of May and minimum (22.4°C) in January. The maximum effluent temperature was observed at the site S_1 (1.0 km from the origin) and minimum at the site S_3 (7.0 km from the origin).

Visibly, the effluent had a dark brown to yellowish brown appearance. The colour of the effluent decreased markedly with increasing distance. The mean value of colour content for site S_1, S_2 and S_3 was 2253.95, 1325.95 and 946.28 CU respectively. Among all the three sites, maximum colour (4019.18 CU) was recorded in the month of October (at site S_1) while minimum (506.55 CU) in the month of August (at site S_3). At site S_1, minimum colour (1427.56 CU) was in the month of March, at site S_2 (935.63 CU) in the month of April and at site S_3 (506.55 CU) in the month of August (Table 1).

The mean value of electrical conductivity for site S_1, S_2 and S_3 was 1.924, 1.302 and 1.034 mS cm^{-1}, respectively. Maximum EC (3.190 mS cm^{-1}) was measured in the month of November and minimum (0.785 mS cm^{-1}) in the month of August. In every month, the electrical conductivity was maximum at the first site S_1 (3.190-1.367 mS cm^{-1}) and minimum at the third site S_3 (1.471-0.785 mS cm^{-1}). There was not dissolved oxygen (DO=0.0 mgl^{-1}) in the effluent samples of all the three sites in every month (Table 1).This may be due to heavy pollution load and microbial activity which have consumed all the oxygen present in the effluent.

Table 2 summarizes the monthly variations among the chemical characteristics of pulp and paper mill effluents at the three sites (S_1, S_2 and S_3). With increasing distance, pH showed a slight shift towards neutrality to mild alkalinity. The mean value of pH for site S_1, S_2 and S_3 was 6.6, 7.0 and 7.2, respectively. In respect of monthly variations, minimum pH (most acidic) was in the months of March and April (5.9) and maximum (alkaline) in the month of June (7.7).

The mean value of lignin content for site S_1, S_2 and S_3 was 4830.18, 3186.87 and 2298.97 mgl^{-1}, respectively. Considering all the three sites, maximum lignin content (5470.99 mgl^{-1}) was at the S_1 site in the month of October and minimum (797.73 mgl^{-1}) in the month of August at the site S_3 showing similar trend as in case of colour. Thus, probability of lignin to colour interdependence may be very high.

The mean value of total phenols for site S_1, S_2 and S_3 was 0.62, 0.27 and 0.12 mgl^{-1}, respectively. Considering all the three sites, the total phenol content was maximum (0.93 mgl^{-1}) in the month of November and minimum (0.03 mgl^{-1}) in the month of April. The concentration of total phenols in the effluent samples was maximum at site S_1 and decreased with increasing distance (Table 2). The mean chloride (Cl^-) content for site S_1, S_2 and S_3 was 240.3, 168.8 and 116.5 mgl^{-1}, respectively. Chloride content decreased with increase in distance from origin. It was maximum (283.3 mgl^{-1}) in the month of November and minimum (69.7 mgl^{-1}) in the month of April. The mean BOD_5 value for site S_1, S_2 and S_3 was 554, 210 and 147 mgl^{-1}, respectively. BOD_5^{20} (Biochemical oxygan demand at 20°C for 5 days incubation) was maximum (686 mgl^{-1}) in the month of November and minimum (97.0 mgl^{-1}) in the month of April. BOD_5^{20} was maximum at the first site (S_1) and decreased with increasing distance from the origin and attaining minimum value at the third site (S_3). The mean chemical oxygen demand for site S_1, S_2 and S_3 was 1317.0, 638.6 and 224.0 mgl^{-1}, respectively. COD was maximum (2057.4 mgl^{-1}) for site S_1 in the month of November and minimum (102.9 mgl^{-1}) for site S_3 in the month of April. In general, COD decreased with increasing distance of effluent stream from the point source.

Data presented in Table 1 and 2 reveals that temperature and pH of the effluent in all the months as well as mean values were below the permissible limits (40°C and 6.5-8.5, respectively) for discharge of effluents in inland surface waters as well as on land for irrigation. Dissolved oxygen was found to be nil and therefore below permissible limit in all the months at all the three sites which indicated a high pollution load (high BOD) in the effluent. The effluent had a dark brown colour at the origin, which

Table - 1: Physical characteristics of effluent from Century pulp and paper mill, Lalkuan (Distt. Nainital) collected at three sites* (S_1, S_2 and S_3) at monthly intervals**

Parameters →	Temperature (°C)						Colour (CU)			Electrical conductivity (mS cm^{-1})			Dissolved oxygen (mg l^{-1})		
Month ↓	S_1		S_2		S_3		S_1	S_2	S_3	S_1	S_2	S_3	S_1	S_2	S_3
	Atm.	Eff.	Atm.	Eff.	Atm.	Eff.									
September	29.9	35.4	27.4	30.5	27.9	29.6	1866.16	979.80	796.7	1.883	1.314	1.077	0.0	0.0	0.0
	±0.0	±0.1	±0.1	±0.0	±0.0	±0.1	±9.53	±9.53	±2.19	±0.025	±0.004	±0.006	±0.0	±0.0	±0.0
October	29.3	34.3	26.9	28.9	26.1	27.7	4019.18	1921.72	1198.23	3.024	1.543	1.252	0.0	0.0	0.0
	±0.1	±0.0	±0.1	±0.0	±0.1	±0.1	±10.30	±8.75	±8.75	±0.066	±0.008	±0.008	±0.0	±0.0	±0.0
November	27.2	32.6	24.2	26.2	22.8	23.6	2224.75	1405.30	1021.47	3.190	1.986	1.471	0.0	0.0	0.0
	±0.0	±0.1	±0.1	±0.1	±0.1	±0.1	±7.88	±3.78	±4.38	±0.011	±0.007	±0.010	±0.0	±0.0	±0.0
December	14.8	30.8	16.2	25.1	15.1	23.3	2017.68	1195.71	828.29	1.600	1.128	0.850	0.0	0.0	0.0
	±0.1	±0.0	±0.1	±0.1	±0.1	±0.0	±5.79	±5.79	±5.79	±0.004	±0.004	±0.003	±0.0	±0.0	±0.0
January	21.60	28.6	18.7	23.1	20.4	22.4	2446.19	1597.84	1112.24	1.750	1.179	0.928	0.0	0.0	0.0
	±0.0	±0.0	±0.0	±0.0	±0.0	±0.0	±5.64	±5.12	±1.38	±0.012	±0.015	±0.005	±0.0	±0.0	±0.0
February	24.4	31.3	22.3	26.7	23.4	24.5	2431.45	1734.93	1111.81	1.920	1.382	1.118	0.0	0.0	0.0
	±0.0	±0.0	±0.1	±0.2	±0.0	±0.0	±7.54	±4.26	±5.50	±0.011	±0.007	±0.008	±0.0	±0.0	±0.0
March	27.0	33.7	27.8	29.1	28.1	27.8	1427.56	1224.69	946.36	1.838	1.309	1.063	0.0	0.0	0.0
	±0.0	±0.0	±0.0	±0.0	±0.0	±0.0	±6.25	±5.32	±1.27	±0.016	±0.005	±0.006	±0.0	±0.0	±0.0
April	36.5	32.3	35.2	31.8	35.1	28.1	1452.83	935.63	805.47	805.47	1.035	0.863	0.0	0.0	0.0
	±0.1	±0.1	±0.0	±0.0	±0.1	±0.1	±9.85	±4.74	±4.22	±0.012	±0.004	±0.002	±0.0	±0.0	±0.0
May	39.4	35.8	36.2	31.5	40.2	30.8	1898.36	1116.22	864.04	1.515	1.128	0.962	0.0	0.0	0.0
	±0.1	±0.0	±0.0	±0.0	±0.0	±0.0	±7.46	±4.42	±3.36	±0.007	±0.008	±0.002	±0.0	±0.0	±0.0
June	41.6	33.2	38.7	32.8	41.6	32.4	2864.52	1566.37	1257.46	1.569	1.176	0.998	0.0	0.0	0.0
	±0.1	±0.0	±0.0	±0.0	±0.0	±0.0	±10.68	±5.58	±5.90	±0.010	±0.002	±0.001	±0.0	±0.0	±0.0
July	33.9	34.8	31.4	31.0	34.4	30.3	2564.41	1247.89	906.76	1.653	1.294	1.046	0.0	0.0	0.0
	±0.0	±0.0	±0.01	±0.0	±0.1	±0.0	±3.82	±5.76	±5.89	±0.003	±0.003	±0.002	±0.0	±0.0	±0.0
August	32.4	33.7	27.9	28.9	27.6	26.9	1834.30	985.33	506.55	1.786	1.145	0.785	0.0	0.0	0.0
	±0.2	±0.0	±0.1	±0.0	±0.0	±0.0	±4.83	±3.77	±1.78	±0.004	±0.002	±0.001	±0.0	±0.0	±0.0
Mean	29.8	33.0	27.7	28.8	28.6	27.3	2253.95	1325.95	946.28	1.924	1.302	1.034	0.0	0.0	0.0
	±7.3	±2.0	±6.6	±2.8	±7.6	±3.1	±676.26	±306.28	±199.73	±0.552	±0.245	±0.180	±0.0	±0.0	±0.0

* S_1 1.0 km from origin
S_2 6.0 km from origin
S_3 7.0 km from origin

Atm. Atmospheric temperature
Eff. Effluent temperature
± Standard deviation

** Mean of two replicates

Table - 2: Chemical characteristics of effluent from Century pulp and paper mill, Lalkuan (Distt. Nainital) collected at three sites* (S_1, S_2 and S_3) at monthly intervals**

Parameters→ Month↓	pH			Lignin concentration (mg l^{-1})			Total phenols (mg l^{-1})			BOD (mg l^{-1})			Chloride content (mg l^{-1})			COD (mg l^{-1})		
	S_1	S_2	S_3	S_1	S_2	S_3	S_1	S_2	S_3	S_1	S_2	S_3	S_1	S_2	S_3	S_1	S_2	S_3
September	7.4	7.5	7.6	4499.32	2539.81	1529.01	0.68	0.27	0.10	556	221	147	235	166	107	1204.0	566.1	168.3
	±0.0	±0.0	±0.0	±4.68	±8.43	±6.18	±0.2	±0.01	±0.00	±14	±12	±15	±4.2	±2.8	±1.4	±9.8	±7.2	±7.2
October	7.1	7.2	7.6	5470.99	4168.69	2731.44	0.71	0.31	0.15	588	228	182	244	186	132	1242.6	657.4	300.2
	±0.0	±0.0	±0.0	±6.14	±2.34	±2.34	±0.01	±0.0.01	±0.01	±20	±16	±17	±2.8	±2.8	±0.0	±5.4	±5.4	±0.0
November	6.5	7.0	7.3	5091.77	3861.00	2511.47	0.93	0.39	0.22	686	274	221	283.3	203.9	163.7	2057.4	981.8	395.9
	±0.0	±0.0	±0.0	±10.19	±8.43	±2.34	±0.02	±0.2	±0.01	±16	±13	±12	±4.0	±4.0	±1.3	±15.8	±5.7	±0.0
December	6.9	7.2	7.4	5043.18	3626.18	2480.43	0.42	0.18	0.07	528	178	112	232.6	159.9	103.4	1157.1	535.9	150.5
	±0.0	±0.0	±0.0	±6.18	±4.68	±2.34	±0.01	±0.01	±0.00	±16	±7	±11	±4.1	±1.3	±0.0	±5.8	±11.5	±5.4
January	6.7	6.9	7.0	5049.99	3927.74	3045.81	0.46	0.25	0.13	578	226	170	239.0	163.6	111.4	1234.7	636.3	290.4
	±0.0	±0.0	±0.0	±7.42	±3.95	±3.43	±0.01	±0.03	±0.00	±11	±10	±11	±2.5	±3.2	±1.0	±6.6	±5.1	±2.3
February	6.3	6.9	7.3	4970.37	3730.43	2544.48	0.82	0.37	0.22	678	260	217	272.6	201.0	161.7	1912.8	974.3	387.0
	±0.0	±0.0	±0.0	±9.73	±8.17	±5.42	±0.04	±0.02	±0.02	±14	±13	±13	±3.8	±3.2	±3.3	±11.7	±8.5	±6.1
March	5.9	7.0	7.2	4171.42	2989.73	0.63	0.63	0.26	0.05	529	186	108	229.8	163.3	97.2	1158.3	560.4	149.1
	±0.0	±0.0	±0.0	±7.16	±4.63	±0.03	±0.03	±0.00	±0.00	±12	±6	±9	±1.6	±4.31	±1.0	±8.6	±6.2	±1.2
April	5.9	6.7	6.8	4249.52	2024.43	1580.01	0.40	0.16	0.03	355	154	154	212.0	131.7	69.7	1023.2	423.7	102.9
	±0.0	±0.0	±0.0	±8.43	±3.98	±2.84	±0.03	±0.02	±0.00	±6	±11	±7	±1.3	±2.1	±0.0	±5.9	±3.3	±0.0
May	6.1	6.5	6.6	4647.25	2593.07	2242.10	0.51	0.27	0.11	513	179	113	222.7	166.7	108.2	1140.4	544.8	154.5
	±0.0	±0.0	±0.0	±5.29	±2.51	±3.46	±0.03	±0.02	±0.01	±6	±10	±8	±3.7	±3.7	±0.0	±10.5	±5.5	±3.8
June	6.6	7.4	7.7	5140.22	3719.86	3183.05	0.82	0.33	0.18	593	242	184	259.9	176.3	1451.1	1355.3	697.0	309.8
	±0.0	±0.0	±0.0	±2.86	±5.83	±2.91	±0.02	±0.00	±0.00	±12	±13	±16	±4.7	±5.2	±1.3	±9.5	±6.1	±8.3
July	6.6	6.8	6.9	5120.22	3015.79	2449.41	0.41	0.17	0.05	517	181	104	222.5	142.0	92.4	1143.9	530.8	139.1
	±0.0	±0.0	±0.0	±6.39	±2.78±	±1.59	±0.00	±0.02	±0.00	±10	±6	±13	±3.3	±2.3	±1.2	±8.3	±5.4	±2.8
August	6.6	6.9	7.1	4507.91	2045.67	797.73	0.64	0.28	0.09	531	186	113	230.6	165.8	106.0	1174.8	554.7	150.7
	±0.0	±0.0	±0.0	±5.31	±3.33	±4.20	±0.01	±0.01	±0.00	±12	±12	±9	±1.9	±2.1	±0.0	±4.9	±6.5	±3.0
Mean	6.6	7.0	7.2	4830.18	3186.87	2298.97	0.62	0.27	0.12	554	210	147	240.3	168.8	116.5	1317.0	638.6	224.0
	±0.4	±0.3	±0.3	±386.28	±720.70	±651.38	±0.17	±0.07	±0.06	±82	±36	±44	±20.4	±20.2	±27.3	±309.3	±165.8	±100.0

* S_1 1.0 km from origin
S_2 6.0 km from origin
S_3 7.0 km from origin

± Standard deviation
** Mean of two replicates

gradually turned in light brown to pale colour with increasing distance from the origin. However, the colour of effluents in all the months was much above the minimum national standards (MINAS). This specifies that the industry should make all efforts to remove colour and unpleasant odour as far as possible, before the discharge of the effluent into environment (Shukla, 1989). Biochemical oxygen demand (BOD_5) of the effluent, a measure of biodegradable organic matter, was found to be much higher in all the months than the permissible limits for inland surface waters (30 mgl^{-1}) as well as for land irrigation (100 mgl^{-1}). Chemical oxygen demand (COD), which is a measure of the oxygen required to completely oxidise the organic carbon, was also found to be much above than the permissible limits (250 mgl^{-1} for both inland surface water and land irrigation) for site S_1 and S_2. However, at site S_3 the mean COD value (224 mgl^{-1}) was found to be below permissible limits. Total phenols in the effluent were below the permissible limit (1.0 mgl^{-1}) in all the months. However, lignin concentration was found to be very high in all the months. Chloride content was below the permissible limits both for inland surface water (600 mgl^{-1}) and for land irrigation (250 mgl^{-1}). Thus, the mean values of temperature, pH, chlorides and total phenols of the effluent were within the permissible limits whereas colour, BOD_5, COD and lignin concentration were found to be above the permissible discharge limits for the industrial waste waters.

The values for various physicochemical characteristics obtained in the present study are in good agreement with the characteristics of pulp and paper mill effluents as reported by other workers. Abbasi (1985) reported that in the combined waste water, pH ranged from 6.5 to 8.2. Singh *et al.* (1996) reported pH range of 6.9 to 8.6 in the combined effluents of Shreyans paper mills Ltd., Ahmedgarh, Punjab. Similarly, most workers have reported the colour of the discharged effluent was dark brown to light brown (Mahajan, 1985; Singh *et al.*, 1996). Mean BOD and COD values obtained in the present study ranged from 147 (site S_3) to 554 (site S_1) mgl^{-1} and 224 (site S_3) to 1317 (site S_1) mgl^{-1}, respectively. These values are higher than the range of 75 to 145 mgl^{-1} and 595 to 800 mgl^{-1} for BOD and COD, respectively as reported by Singh *et al.* (1996), but lower than the range of 1286 to 1764 mgl^{-1} and 2653 to 3369 mgl^{-1} for BOD and COD, respectively as reported by Chakravarthi *et al.* (1995) for Shri R.R. paper mill, Nuzvid. These differences may be due to variations in manufacturing processes, production capacity and efficiency of in-mill treatment plants as well as sites of effluent collection.

Data on monthly variations (Table 1 and 2) in effluent characteristics (*e.g.* colour, lignin, BOD, total phenols, chlorides and COD) revealed that in most cases, the values were higher from October to January, compared to other months. These high values were accompanied by low atmospheric and effluent temperatures in these months. Singh *et al.* (1996) found that the colour and COD values were maximum in the months of October to November. Sahoo *et al.* (1997) also found high values of COD in the months of September to February for Modi cement factory, Madhya Pradesh. Thus, it may be concluded that the high values for these pollution parameters reflect the reduced activity of indigenous micro flora present in the effluent, responsible for natural bioremediation.

Data on the effect of spatial changes on effluent quality parameters were recorded at long range distances (Table 1, 2) in different months. In all cases, it was observed that there was a continuous decline in both physical characteristics as well as chemical characteristics in all cases. The decline in BOD, COD and total phenol values was almost four to five folds, whereas it was about two to three folds in case of lignin and chloride concentrations. The changes in the effluent characteristics with increasing distance suggest that the effluent undergoes the process of self purification, which may be due to synergistic action of indigenous microorganisms, aeration, pH changes, photochemical effects and dilution. Archibald *et al.* (1998) found that photolysis plays an important role in mineralization of organic pollutants from the biologically treated pulp and paper mill effluents.

The present study on significant changes in effluent quality parameters with the seasonal and spatial distribution emphasizes the need for using identical site and season for comparing the effluent characteristics, as has also been emphasized by the results of Singh *et al.* (1996). The data bank generated by monthly monitoring studies of paper mill effluent could be successfully used in prediction of tedious and time consuming parameters by laboratory assessment of easily measurable simple parameters (Malaviya and Rathore, 2001).

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