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Research Article

# Effluents from Paper and Pulp Industries and their impact on soil properties and chemical composition of plants in Uttarakhand, India

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**Industrial effluents of Paper and Pulp industries Mill were characterized and their impact on soil properties and chemical composition of crop plants was studied in Uttarakhand State of India. The effluent characteristics particularly, total suspended and total dissolved solids, biological oxygen demand, chemical oxygen demand and the content of Na were higher than the prescribed limits. In general, a considerable decrease in the concentration of total suspended and dissolved solids, bicarbonate, chloride, ammoniacal, calcium, sodium, potassium, copper, manganese and cobalt occurred during the flow of effluents downstream. The lateral seepage of paper and pulp industrial effluent to agricultural lands increased the soil pH and electrical conductivity (EC). At shorter distances from point source, the lateral seepage of effluent increased exchangeable K, Na and extractable S, Zn, Fe, Mn, Pb and Ni in soil. The contents of P, K, micronutrients, Pb and Ni in wheat plants increased due to seepage of effluent to the nearby agricultural fields close to the point source.**

Key words: Effluents, crop plants, nutrients, toxic metals, Paper and pulp industries

## INTRODUCTION

Among the various industries, paper and pulp industry is one of the notorious polluters of the environment. It has been categorized as one of the most polluting industries due to discharge of huge volumes of highly colored and toxic waste water (effluent) in the environment causing pollution of land (soil), air and water (Martin, 1998). Most of the paper and pulp industries discharge their insufficiently treated waste water into the river or stream which results in serious problems for aquatic life (Kesalkar et al., 2012)

The most important problem which the pulp and paper industry is facing today is the disposal of tremendous volumes of waste water. This waste water is rich in dissolved solids such as chlorides and sulphates of Na, Ca and varying amounts of suspended organic materials. In addition to these constituents, effluents also contain some trace metals like Hg, Pb, and Cr etc. The effluents are generally alkaline in reaction with high chemical and biological oxygen demands. Thus, the effluents discharge into the water systems make the water unfit for irrigation and potable use and create health hazards.

Treated industrial waste water could be used safely and effectively with proper precautions to increase the soil productivity (Chhonkar *et al*., 2000). However, despite being a useful source of plant nutrients (N, P, K, Ca etc.), the paper mill effluent often contains high amounts of

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various organic and inorganic materials, as well as toxic trace elements, which may accumulate in soils in excessive quantities under long term use. Subsequently, these toxic elements may cause severe problems to human beings and animals by entering into the food chains. Untreated industrial effluents contain higher amounts of Cd, Pb, Zn, Cu, Mn and Fe and enhance the concentration of the heavy metals in irrigated surface soils (Xiog *et al.,* 2001). Significantly higher values of EC, organic carbon, available K, exchangeable cations ( $Ca^{2+}$, $Mg^{2+}$), exchangeable anion ( $Cl^-$, $HCO_3^-$) along with micro-nutrient cation ($Cu^{2+}$) have been reported in soils being irrigated by paper and pulp industry effluents (Singh et al. 2007). Similarly a decrease in germination percentage, seedling growth and their dry weight in different plants with an increase in paper mill effluent concentration had also been reported (Sundaramoorthy and Kunjithapatham, 2000). Zwieten et al. (2010) reported that the biochar formed due to slow pyrolysis of paper effluent affected the agronomic performance and soil fertility of some soils. Kumar et al. (2010) performed studies to determine the agronomical characteristics of *Trigonella foenum-graecum* irrigated with different concentrations of Paper mill effluent. Their study revealed that the effluent was rich in some plant nutrients and affected the agronomical characteristics of *T. foenum-graecum* (cv. Pusa early bunching) and physico-chemical characteristics of the soil as well.

The present investigation was undertaken with the objective to examine the characteristics of effluents from Paper and Pulp industry and to study the impact of lateral seepage on soil properties of agricultural fields adjoining main drains and changes in the elemental composition of plants therein.

## MATERIALS AND METHODS

### Collection of effluent, soil and plant samples

The paper mill effluent samples were collected in the month of March from four locations downstream the main drain of Century Paper and Pulp Mill, Lalkuan. The samples were collected at a distance of about 0.5 Km ($L_1$), 3.0 Km ($L_2$), 7 Km ($L_3$) and 11 Km ($L_4$) from the discharge point. Two effluent samples were collected in the month of March from Cheema Paper Mill, Bajpur at a distance of 0.5($L_5$) and 1Km ($L_6$) from the discharge point. For comparison purpose, one sample of tubewell irrigation water was collected from nearby unpolluted site located in Pantnagar. The effluent samples were collected in 2 litre plastic cans and were stored in a deep freeze to avoid any degradation prior to chemical analysis.

In order to study the effect of lateral seepage of effluent on soil properties, at each sampling site of Century Paper and Pulp Mill (Lalkuan, India) surface (0-15 cm) soil samples were collected with the help of bucket auger at 0.5 to 1 m or 10 to 15 m laterally away from the effluent drain and were designated as L and LC, respectively.

In order to study the effect of effluent or tube well irrigation water on crop plants, wheat plant samples were also collected from each soil sampling site of Century Paper and Pulp Mill (Lalkuan, India).

### Processing of soil and plant samples

Soil samples were air-dried in shade and crushed using a wooden roller and passed through a sieve having openings of 2 mm diameter. Processed soil samples were stored in plastic bags until analyzed.

Plant samples were washed thoroughly with running tap water to remove the dust. They were then washed in 0.1 *N* HCl solution and finely in distilled water. After air drying, plant samples were dried in an electric oven at a $60^0$ C. The dried plant samples were finely ground in a Wiley mill and stored in plastic packets.

### Chemical analysis of effluents, soil and plant samples

Effluent samples collected from different locations were analyzed for pH, electrical conductivity (EC), Total Solids (TS), Total Dissolved Solids (TDS), Total suspended Solids (TSS), Biological Oxygen Demand (BOD) and Chemical Oxygen Demand (COD), ammoniacal and nitrate nitrogen (N), phosphorus (P), potassium (K), calcium (Ca), magnesium (Mg), sodium (Na), sulphate, carbonates and bicarbonates, chloride, micronutrient cations (Fe, Cu, Mn and Zn) and heavy metals (Co, Cd, Ni, Pb and Cr) as per the standard procedures (APHA, 1998).

Soil samples were analyzed for pH and EC in 1:2 soil water suspension, organic carbon by titrimetry, alkaline $KMnO_4$ hydrolysable N by titrimetry, Olsen's P by colorimetry, 1 N ammonium acetate (pH=7.0) extractable K by flame photometry, diethylene triamine pentaacetic acid (pH 7.3) extractable Zn, Cu, Fe, Mn, Co, Cd, Ni, Pb and Cr content by atomic absorption spectrophotometry as per the procedures outlined by Tandon (1993).

Finely ground plant samples were digested in diacid mixture ($HNO_3$: $HClO_4$, 3:1 v/v) and digests were analyzed for P, S, K, Zn, Cu, Fe, Mn, Co, Cd, Ni, Pb and Cr contents using the techniques mentioned above.

## RESULTS AND DISCUSSION

### Characteristics of industrial effluents

The properties of paper and pulp industry effluents and tube well water are presented in Table 1. The pH of the

**Table 1.** Some characteristics of paper and pulp industry effluents at different locations downstream the main drain.

| Character | Century Paper & Pulp Mill, Lalkuan | | | | Cheema paper mill ,Bajpur | | Tube well water | Tolerance limits for irrigation* |
|---|---|---|---|---|---|---|---|---|
| Location | L1 | L2 | L3 | L4 | L5 | L6 | L8 | |
| pH | 7.32 | 7.38 | 7.46 | 7.65 | 7.48 | 7.56 | 7.58 | 5.5-9.0[a] |
| EC(ds/m) | 1.29 | 1.74 | 1.48 | 0.79 | 1.1 | 1.3 | 0.4 | 2.0-10.0[b] |
| BOD( mg/l) | 306 | 408 | 306 | 306 | 306 | 408 | 51 | 100[a] |
| COD( mg/l) | 4357 | 3983 | 1736 | 647 | 1532 | 1123 | 238 | 250[a] |
| TSS(mg/l) | 460 | 240 | 200 | 80 | 480 | 340 | traces | 200[a] |
| TDS( mg/l) | 1380 | 1120 | 1000 | 480 | 700 | 760 | 160 | 1000[a] |
| TS (mg/l) | 1840 | 1360 | 1200 | 560 | 1180 | 1100 | 160 | - |

[a] CPCB (1975), [b] Paliwal and Yadav (1976)

effluent samples from Century Paper Mill varied from 7.32 to 7.65. The lowest value was recorded at 0.5 km ($L_1$) while the highest value was at farthest location (11 km from Century Paper and Pulp Mill, $L_4$). The pH values of the effluent samples from Cheema Paper Mill varied from 7.48 to 7.56. The lowest value was recorded at 0.5 km ($L_5$) while the highest value was at 1 km ($L_6$) away from paper mill. The increase in pH value of the effluent with distance might be due to dilution with municipal waste water. Considering the Indian Standards limit on pH of effluents (5.5 to 9.0) for their irrigation use (CPCB, 1975), the pH values of all effluent samples fell well within the permissible limit. Similar pH values of the effluents discharged from paper and pulp industry were reported earlier by Kamalakar (1988).

The electrical conductivity of paper mill effluent varied from 0.79 to 1.74 dS m-1 at 25°C. The highest value of EC observed near the source was because of high concentration of dissolved salts which decreased progressively with distance due to dilution as well as adsorption of metal ions on soil colloids or by precipitation reducing the concentration of these ions in the effluent. As Lokhande et al. (1996) reported that an EC value of 3 dS/m or more might affect the germination of most crops, therefore, no effluent sample was likely to pose problems with the seed germination..

The BOD value of effluent varied in the range of 306 to 408 mg L-1 whereas, COD varied in the range of 647 to 4357 mg L-1. The observed values of BOD and COD in the effluents were higher than the recommended value of 100 mg BOD L-1 and 2500 mg COD L-1 as prescribed by CPCB (1975). High COD value might be due to the presence of oxidisable organic compounds in the effluent (Hybes, 1971). The presence of lignin in the waste water which is not easily biodegradable could be responsible for high COD/BOD ratio in these effluents. The COD values also decreased downstream due to settling of some lignin residues in the natural drain.

The values of total solids (TS), total dissolved solids (TDS) and total suspended solids (TSS) in effluents were much higher in comparison to the tube well water and generally decreased with the increasing distance from the outlet. Considering 200 mg total suspended solids L-1 and 1000 mg total dissolved solids L-1 as the threshold values for irrigation use (CPCB, 1975), the effluent water from both paper and pulp industries was not suitable for irrigation, except for the location L4 (480 mg L-1) which was 11 km away from the site of origin.

The concentrations of soluble cations and anions in effluent and tubewell water are presented in Table 2. Among anions, the concentration of bicarbonate and chloride ions in the effluents regularly decreased with the distance except at Bajpur where the concentration of chloride ion was higher even at the farther location possibly due to additional of municipal waste water. No regular pattern of increase or decrease with the distance was noted in the case of nitrates, phosphates and sulphates. Among cations, the concentrations of NH4, Ca, Na, K, Cu, Mn and Co decreased with the increase in distance for effluent of Century Paper and Pulp industry, Lalkuan possibly due to their dilution or precipitation 'however' no such decrease was noted for

**Table 2.** The concentrations of some cations and anions in paper and pulp industry effluents at different locations downstream in the main drain.

| **Content (mg $L^{-1}$)** | **Century Paper & Pulp Mill, Lalkuan** | | | | **Cheema paper mill ,Bajpur** | | **Tube well water** | **Tolerance limits for irrigation** |
|---|---|---|---|---|---|---|---|---|
| **Location** | **L1** | **L2** | **L3** | **L4** | **L5** | **L6** | **L8** | |
| Bicarbonate | 440 | 408 | 396 | 352 | 440 | 515 | 201 | - |
| Chloride | 347.6 | 268 | 195.5 | 43.5 | 57.9 | 72.4 | 7.2 | 600[a] |
| Nitrate -N | 2.48 | 4.96 | 4.96 | 42.16 | 12.4 | 4.96 | 2.48 | - |
| Phosphate-P | 0.98 | 0.46 | 0.58 | 0.80 | 0.22 | 0.26 | 0.08 | - |
| Sulfate- S | 0.27 | 0.36 | 0.15 | 0.17 | 0.26 | 0.48 | 0.07 | 1000[a] |
| Ammonical- N | 7.92 | 6.48 | 5.04 | 0.72 | 1.44 | 0.72 | 0 | - |
| Calcium | 2346 | 2080 | 2195 | 1614 | 1954 | 2261 | 1336 | - |
| Magnesium | 51 | 51 | 51 | 50 | 68 | 88 | 41 | - |
| Sodium | 826 | 807 | 817 | 347 | 450 | 758 | 13 | 60[a] |
| Potassium | 334 | 270 | 240 | 128 | 111 | 166 | 62 | - |
| Iron | 0.73 | 0.33 | 0.45 | 0.17 | 0.32 | 1.04 | 0.33 | 5.0-20.0[c] |
| Copper | 0.006 | 0.0005 | 0.0005 | 0.0003 | 0.002 | 0.001 | 0.001 | 0.2-5.0[c] |
| Manganese | 0.25 | 0.23 | 0.14 | 0.07 | 0.17 | 0.23 | 0.001 | 0.2-10.0[c] |
| Zinc | 0.04 | 0.006 | 0.01 | 0.012 | 0.02 | 0.029 | 0.03 | 2.0-10.0[c] |
| Cobalt | 0.021 | 0.007 | 0.011 | 0.004 | 0.012 | 0.008 | ND | - |
| Cadmium | 0.004 | ND | ND | ND | ND | ND | ND | 0.01-0.05[c] |
| Lead | 0.081 | 0.045 | 0.067 | 0.05 | 0.06 | 0.071 | 0.053 | 5.0-10.0[c] |
| Chromium | 0.007 | ND | ND | ND | ND | ND | ND | 0.1-1.0[c] |
| Nickel | 0.019 | 0.009 | 0.015 | 0.01 | 0.009 | 0.012 | 0.005 | 0.2-2.0[c] |

[a] CPCB (1975), [b] Paliwal and Yadav (1976), [c]Ayers and Westcot (1976)

Cheema Paper and Pulp industry effluent, Bajpur owing to the shorter distance of observation and possible addition of some municipal waste water in the effluent. Rest of the cations (Mg,, Fe, Zn, Cd, Pb, Ni and Cr ) did not show any regular pattern of increase or decrease in their concentrations. The concentration of Na in effluents at all sites was much higher than the permissible limit (60 mg L-1) while the concentrations of other observed cations or anions were present in concentrations lower than their permissible limit for use in irrigation.

**Soil properties at different lateral distance from the main drain**

Some properties of soil samples drawn at different lateral distance from the main drain are shown in Table 3. It is apparent from the data that the soil sample adjacent to the main drain had higher value of pH and electrical conductance than the soil lying much away from the effluent drain which could be attributed to the lateral seepage of effluent from the unlined drain. However, at $L_3$ site the soil pH near the drain was lesser than pH of the soil lying to 10- 15 m away from the drain.

At the closest sampling point (0.5 km from discharge point), soil sample drawn within 0.5 m away from the main drain had the higher contents of only soil exchangeable K and Na, 0.15% CaCl2 extractable S and DTPA extractable Zn, Fe, Mn, Pb and Ni as compared to the soil sample drawn 15 m laterally away. However, no trend was noted for other sampling points which were much distant (3 and 7 km) from the source point. The differences in the

**Table 3.** Some properties of soil samples collected at varying distance from the main effluent drain

| Properties | Locations (km from discharge point) | | | | | |
|---|---|---|---|---|---|---|
| | 0.5 | | 3 | | 7 | |
| | L1 | L1C | L2 | L2C | L3 | L3C |
| pH (1:2) | 8.76 | 8.49 | 7.50 | 7.10 | 7.83 | 8.10 |
| EC (1:2) in ds $m^{-1}$ | 0.38 | 0.27 | 0.3 | 0.22 | 0.35 | 0.32 |
| Organic C (g $kg^{-1}$) | 12.0 | 13.9 | 14.8 | 12.3 | 16.7 | 19.6 |
| Alk. KMnO4 hydrolysable N (mg $kg^{-1}$) | 71.4 | 107.8 | 127.4 | 137.2 | 131.6 | 140.0 |
| Olsen's P (mg $kg^{-1}$) | 16.1 | 17.5 | 10.2 | 3.9 | 26.5 | 16.6 |
| 1M NH4OAC extr. K (mg $kg^{-1}$) | 45.5 | 42.7 | 36.0 | 109.5 | 53.0 | 43.0 |
| 1M NH4OAC extr. Na (mg $kg^{-1}$) | 184.0 | 126.5 | 101.2 | 126.5 | 154.1 | 184.0 |
| 0.15% CaCl2 extr. S (mg $kg^{-1}$) | 119.9 | 52.3 | 114.2 | 52.8 | 97.0 | 114.2 |
| DTPA extr. Zn (mg $kg^{-1}$) | 1.8 | 0.8 | 0.7 | 1.2 | 1.9 | 1.7 |
| DTPA extr. Cu (mg $kg^{-1}$) | 1.8 | 1.7 | 1.8 | 2.4 | 2.5 | **2.7** |
| DTPA extr. Fe (mg $kg^{-1}$) | 19.2 | 15.0 | 14.4 | 48.2 | 38.0 | 40.8 |
| DTPA extr. Mn (mg $kg^{-1}$) | 7.8 | 6.1 | 8.2 | 29.3 | 10.3 | 11.2 |
| DTPA extr. Co (mg $kg^{-1}$) | ND | ND | 0.1 | ND | ND | ND |
| DTPA extr. Pb (mg $kg^{-1}$) | 1.7 | 0.8 | 1.1 | 1.2 | 2.1 | 2.0 |
| DTPA extr. Ni(mg $kg^{-1}$) | 0.10 | 0.08 | 0.09 | 0.31 | 0.14 | 0.13 |
| DTPA extr. Cd (mg $kg^{-1}$) | 0.01 | ND | 0.01 | 0.02 | 0.01 | 0.01 |

**Table 4.** Elemental composition of wheat plants collected at varying distances from the main effluent drain

| Content (mg $kg^{-1}$) | Locations (km from discharge point) | | | | | |
|---|---|---|---|---|---|---|
| | 0.5 | | 3 | | 7 | |
| | L1 | L1C | L2 | L2C | L3 | L3C |
| P | 2985 | 2674 | 2130 | 1889 | 1948 | 2038 |
| K | 18413 | 14825 | 13950 | 6038 | 862.5 | 12425 |
| Na | 225 | 325 | 150 | 125 | 88 | 113 |
| Ca | 1284 | 1374 | 1319 | 2864 | 1606 | 1438 |
| Mg | 811 | 1339 | 130 | 731 | 173 | 60 |
| S | 1600 | 1600 | 4000 | 1600 | 800 | 1000 |
| Zn | 17 | 14 | 22 | 19 | 12 | 14 |
| Cu | 3.1 | 2 | 4.6 | 2.6 | 1.1 | 1.4 |
| Fe | 93 | 74 | 119 | 29 | 39 | 40 |
| Mn | 20.0 | 12.8 | 33.1 | 25.2 | 8.7 | 12.8 |
| Co | ND | ND | ND | ND | ND | ND |
| Pb | 4.1 | 3.7 | 6.8 | 2.8 | 2.5 | 2.7 |
| Ni | 0.75 | ND | 0.1 | ND | ND | ND |
| Cd | ND | ND | ND | ND | ND | ND |

content of rest soil extractable nutrients/pollutants were not much. Earlier reports (Achari et al., 1999 and Singh, 2007) also confirmed a significant increase in soil EC, exchangeable cations and micronutrient cations in the soil irrigated by the effluent of paper mill.

### Effect of lateral seepage of paper mill effluent on elemental composition of crop plants

The elemental composition of wheat plants sampled at different lateral distances from the main drain at different sampling points is presented in Table 4. It is apparent from the data that wheat plants growing within 0.5 m away from the unlined drain had much higher concentrations of P, K, Zn, Cu, Fe, Mn, Pb and Ni as compared to the plants which were collected 15 m away from the main drain. The effect could be ascribed to the lateral seepage of effluent from the unlined main drain. However, no such trend was evident at the farther sampling point (7 km from the point source).

## CONCLUSION

Thus, it can be concluded that the paper and pulp industrial effluent characteristics particularly, the total suspended and soluble solids, BOD, COD and content of Na were higher than the prescribed limits. In general, there is a considerable decrease in the concentration of solids and some soluble nutrients in the paper and pulp industry effluents during the downstream flow. The lateral seepage of effluent increased exchangeable- K, -Na and extractable S, Zn, Fe, Mn, Pb and Ni in surface (0-15 cm) soil and the contents of P, K, micronutrients, Pb and Ni in crop plants, especially, at shorter distances from the point source.

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