EVALUATION OF SOME ENVIRONMENTAL INDICATORS OF TANNERY WASTEWATER IN OSUMI RIVER

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Abstract

A wide range of chemicals and processes are used in leather industry, which have a negative impact on aquatic ecosystem and in the quality of environmental performance. This study aimed to assess some environmental indicators of tannery wastewater in Osumi River. Although it is recognized the pollution of Water River by discharges of leather processing, never before has been conducted a genuine study for assessment of pollution in the area. Water samples have been taken at four important and different places. All samples were analyzed according to standard methods and reagents of high analytical quality. All the physical-chemical indicators of leather wastewater depend from the different phases of leather processing. pH varies at limits 9,2-12,5; Total alkalinity varies from 92-2245 $mg/L\ CaCO_3$; TSS varies from 243-2421.6 mg/L; TDS varies at limits 918-8974,41 mg/L; ammonia ions (NH_4^+) from 3,52-45 mg/L; chlorides (Cl^-) varies at limits 97,725-10280,5 mg/L; phosphates (PO_4^{3-}) varies from 1,2-3,25 mg/L; T^0C varies from $12^0 - 23.5^0C$; chromium (VI) varies at limits 0,1-0,45 mg/L; total Fe from 0,7-2,25 mg/L; values of $COD_{(Mn)}$ varies from 145,6-11200 $mg/L\ O_2$; nitrate ions (NO_3^-) varies from 5,7-45,8 mg/L. This survey provided the latest data on the quality of industrial effluent in leather processing, based on the definition of some environmental contaminants. Composite untreated wastewater has been found turbid, colored and smelling. The Study of Osumi River pollution by leather effluent is an innovation. Related negative impact of leather effluent on the environment, leather wastewater treatment remains a priority for the future.

***Keywords**: environmental indicators, tannery wastewater, Osumi River.*

Introduction

A wide range of chemicals and processes are used in leather industry. Different forms of waste in quality and quantity were produced, which have a negative impact on aquatic system and environment. In Albania as well as in many developing countries, leather processing has been implemented in the private sector. The common characteristic of all tanneries in Albania is their position near the river[6]. They have no treatment plant for leather effluent. This study aimed to assess some environmental indicators of tannery wastewater in Osumi River. Although is recognized river water pollution by discharges of leather processing, before is not carried out a genuine study for the assessment of pollution in the area. Discharge of untreated leather effluent made directly to surface waters Osumi River, and formally exist sedimentation tank. Leather effluent pass through a drainage channel, that extends through agricultural lands, until joining the recipient waters of the river. Leather wastewaters are alkaline, with color and smell extremely unpleasant and irritating, especially to allergic persons and respiratory system. Estimated that for every ton of raw hide/skin, are produced 30-50m³ water discharges and resulting in the production of 100-150 m² finished leather[6], [8]. The amount of wastewater discharge in a tannery depends on used technology, the quality and quantity of raw materials. Distinguished processes are: pickle process and tanning process[8]. Wastewater generated by leather processing may be classified as follows: 1- Wastewater from beam house processes including: (soaking &washing, liming, unhearing, declaiming, bating, degreasing); 2- wastewater from tanning processes including: (pickling, chrome-tanning); 3- wastewater from post-tanning and finishing processes including: (retaining, fat liquoring, coloring & finishing).

Fig.1. Wastewater produced by leather processing.

Materials and methods

Initially was located and determined accurate place of effluent discharge, the way through the drainage channel is joining the river. Water sampling was carried out according to references values[10]. This process is difficult to carry out because of the various phases and periodical discharges according to the quantity and composition. Sampling was carried out at 4 important places: Station 1- Leather effluent drainage channel. Station 2- Place where leather wastewater discharged into river basin. Station 3- River of Vodica, a village about 5 km away from Berati, where is a small tannery. Station 4- Osumi River station, close to the city center. Most parameters such as pH, ammonia ions (NH_4^+), nitrate ions (NO_3^-), phosphates (PO_4^{3-}), $COD_{(I_{Mn})}$, were tested during the day of collection. For other parameters precautions and storage of samples were done according to references values.

Methods for wastewater analyzes

All physic-chemical parameters were analyzed according to standard methods *EN ISO*. *pH* was determined using "Selecta", ISO-9001 certified, pH-meter. Total alkalinity analyzed with titration method[9]. $COD_{(I_{Mn})}$ was determined with standard method of permanganate index[13]. Leather wastewaters are heavily contaminated waters, provided an appropriate pre-dilution is adopted. Ammonia ions (NH_4^+) were determined by nesslerization method[12], Nessler´s reagent. Nitrate ions (NO_3^-) were determined by colorimetric method –Test Kits/2,6 dimethylphenol. Phosphates (PO_4^{3-}) were determined with colorimetric method with ammonium molybdate[11]. Chromium (VI) is determined with photometric Test Kits/reaction with diphenylcarbazide. Iron ions $Fe^{2+/3+}$ were determined with photometric Test Kits. Ions chlorides (Cl^-) were determined with standard method of titration (Argentometria)[1]. *TSS* was determined with filtration and gravimetric method[14]. *TDS* was determined with evaporation, drying and gravimetric method[14].

Results and discussion

Physic-chemical characteristics of leather discharges depend from quantity and quality of raw materials, various types of organic and inorganic reagents, used during different stages of leather processing. Leather wastewater was not carried out any preliminary treatment, before being discharged into river.

Table 1 Indicators of leather wastewater quality (mg/L)

Indicators	Sampling Dates					
	18/4/2012	15/5/2012	18/5/2012	25/6/2012	16/8/2012	18/9/2012
Temp.℃	18	21	21.5	23	23.5	22.5
PH	9.87	10.7	9.5	9.8	10.07	12.5
Total alkalinity (mg/L $CaCO_3$)	264.57	380.9	271.34	476.45	1568	2245
COD (IMn) (mg/L O2)	445.8	326.4	785.2	1143.9	1644.8	11200
Ammonia ions (mg/L)	20.5	25.8	13.5	15	25	35.8
Nitrate ions *(mg/L)*	25	30.8	22.5	35	25.8	45.5
*Phosphate (mg/*L)	1.5	1.8	2.35	1.76	2.18	3.25
Chloride (*mg/L*)	836.5	1047.58	1140	1985	3852	4608.4
TSS (mg/L)	1047.8	1136.2	1631.6	1013.8	1094.5	2167
TDS (mg/L)	1969.4	2180.2	2618	2784.41	3171.8	8974.41
*Chromium (*VI) (mg/L)	0.18	0.15	0.17	0.25	0.3	0.45
Iron ions (mg/L)	1.2	0.9	1.2	1.5	2.2	2.5

Indicators	Sampling Dates					
	23/10/2012	16/11/2012	14/1/2013	28/1/2013	12/2/2013	18/2/2013
Temp.℃	17	16.5	14	15	12	11.5
PH	11.4	11.5	11.7	10.8	9.2	9.3
Total alkalinity $_{(mg/L}$ $CaCO_3)$	1634.2	1896.4	2017.5	1985	89	92
COD $_{(IMn)}$ $_{(mg/L}$ $O_2)$	1540	438.4	327.8	416	145.6	167.2
Ammonia ions $_{(mg/L)}$	22.8	24.5	37	45	3.52	6.5
*Nitrate i*ons	27.8	36.8	52.5	45.8	12	5.7
Phosphate (mg/L)	2.2	1.5	2	2.5	1.2	1.5
Chloride (mg/L)	2341.5	3841.4	6978.5	10280.5	97.725	276.51
TSS (mg/L)	1684	2421.6	1878.4	1059	427	243
TDS (mg/L)	2025.4	2218.2	3261.5	8286	992	918
Chromium (VI) $_{mg/l}$	0.4	0.25	0.3	0.25	0.1	0.1
Iron ions (mg/L)	1.5	0.8	1.0	1.2	0.7	0.8

Table 1 presents that the values of the measured parameters such as pH, Total alkalinity, $COD_{(I_{Mn})}$, chloride, ammonia ions, chromium (VI), TSS; TDS exceed many times the domestic standards. The values of these indicators are lower in the dates 12/2/2013 and 18/2/2013 as a result of dilution process of leather wastewater in a rainy weather conditions.

Table 2 Indicators of leather effluent in recipient water body in Osumi River

Indicators	Sampling Dates					
	27/3/2012	18/4/2012	8/5/2012	25/6/2012	20/8/2012	11/9/2012-
Temp.℃	13	15	16.8	17.1	19.2	18.8
PH	9	8.5	8.2	8.5	9	8.5
Total alk.	274.6	149.23	78.6	240.18	265.85	148.45
COD $_{(IMn)}$	147.14	108.28	76.48	106.86	192.58	164.46
*Ammonia io*ns	20	15	16	20	25	20.5
*Nitrate ion*s	20	15	10	18	22	20
*Phosphat*e	1.25	1.2	0.5	1.2	1.5	1.25
Chloride	103.71	113.41	98.42	117.64	213.72	123.46
TSS	380.52	281.32	202.71	179.87	216.98	209.85
TDS	643.46	583.51	1814.7	2061.7	2739.5	3723.1
*Chro*mium (VI)	0.15	0.1	0.15	0.1	0.2	0.15
Iron ions	1.0	0.5	1.0	0.5	1.5	1.2

Indicators	Sampling Dates					
	18/10/2012	16/11/2012	6/12/2012	14/1/2013	31/1/2013	7/2/2013
Temp. ℃	16.5	14.2	16.5	12.4	11.2	11.5
PH	8.2	8.4	8.5	9.0	9.5	9.8

Total alk.	98.64	87.315	102.34	183.45	1400	1600
COD (IMn)	112.78	98.48	82.95	196.98	264	272
Ammonia ions	15	18	13	15	17	20
Nitrate ions	15	10	15	18	25	32.5
Phosphate	1.5	1.0	1.5	0.8	0.5	1.2
Chloride	65.85	71.64	93.42	216.5	815.35	850.8
TSS	284.45	199.67	237.52	86.9	194	72
TDS	1673.8	1968.1	2162.7	681	2245	2195
Chromium (VI)	0.12	0.15	0.15	0.12	0.15	0.2
Iron ions	1.2	0.8	1.3	1.0	0.5	1.2

Parameters of Table 2 have the measured units as Table 1.Values of the parameters in Table 2 is lower compared with the values of Table 1. Reduction of pollutants from tannery wastewater values has come as a result of dilution process receiving water area of the river.

Table 3. Reduction of pollutants of leather effluent in recipient water body of Osumi River

Parametrat	Station 1 max value	Station 2 max value	Reduction of pollutants %
pH	12.5	9.8	21.59
COD	11200	196.98	98.24
NH_4^+	45	25	44.4
Total alk.	2245	1600	28.73
NO_3^-	45.8	25	48.45
PO^{3-}_4	3.25	1.5	53.84
Cl	10280.5	850.8	91.72
Cr(VI)	0.45	0.2	55.5
TSS	2421.6	380.52	84.28
TDS	8974.41	3723.1	58.51

Table 3 shows the reduction of maximum values of pollutants from Station 1 to Station 2 and this fact indicates the river water pollution in the area by leather wastewater.

Table 4 Physicochemical parameters of the River of Vodica

Indicators	Sampling Dates					
	15/5/2012	25/6/2012	7/8/2012	21/972012	12/10/2012	8/11/2012
Temp.℃	16	19	18	20	11	15
PH	8.5	8.2	8.1	8	7.8	8.2
Total alk.	86	41	28	26	21.4	34
COD (IMn)	9.88	3.2	21.6	8.48	8.6	15.2
Ammonia ions	3.5	2.25	0.15	1.5	1.8	1.5
Nitrate ions	10	10	0.15	10	10	3
Phosphate	0.15	0.075	0.15	0.075	0.1	0.075
Chloride	27.65	21.32	71	27.8	49.63	22.42
TSS	208.46	127	18	90.84	43.71	61.57
TDS	215.8	148.72	81.64	108.65	61.34	89.4

*Chromium (*VI)	0.02	0.01	0.01	0.01	0.01	0.02
Iron ions	0.1	0.05	0.05	0.1	0.1	0.1

Indicators----------Sampling Dates----------

	16/11/2012	11/1/2013	18/1/2013	25/1/2013	1/2/2013	8/2/2013
Temp.℃	15	11.5	11	11	12	11.5
PH	8.4	8.7	8.5	8.2	8.6	8.8
Total alk.	65	107	98	81.92	92	113.5
COD (IMn)	27.6	8.32	7.63	9.86	11.56	47.2
Ammonia ions	1.8	0.8	1.5	1.25	2.5	0.25
Nitrate ions	15	10	8	10.5	15	3.5
Phosphate	0.15	0.15	0.1	0.1	0.15	0.25
Chloride	53.18	21.25	36.4	27.86	57.05	28.35
TSS	73.4	176	142.8	131.12	183.45	714
TDS	92.71	284. 56	206. 8	198.53	275.67	982.78
*Chromium (*VI)	0.02	0.01	0.01	0.02	0.02	0.03
Iron ions	0.02	0.5	0.2	0.1	0.2	0.3

The data in Table 4 show the physic-chemical parameters of Vodica River as part of Osumi River. This area was determined Station 3.It is important because 100-150 m away is a small tannery, that discharges untreated leather wastewater. During the period of sampling this tannery was outside the function or has functioned not periodically. This fact proves the level of contaminants, which is many times lower compared with the values of pollutants in Osumi River basin, where were deposited leather effluent of the tannery in Berati, in the distance 300-350 m off the coast of the river.

Table 5 Physic-chemical parameters of Osumi River in Station 4

Indicators----------Sampling Dates----------

	18/1/2012	26/2/2012	15/3/2012	18/4/2012	8/5/2012	13/6/2012	19/7/2012
Temp.℃	12	11.8	12.5	14	16	19.5	18.5
PH	8	7.8	8.1	8.3	8.5	8.4	8.6
Total alk.	6.5	5.75	8.78	6.5	9.8	8.35	13
COD (IMn)	36.4	38.8	26.24	36.8	32.8	24.6	6.8
*Ammonia ion*s	0.2	0.15	0.22	0.2	0.5	1.5	1.8
Nitrate ions	3.8	9.5	5.5	4.8	12	13.7	10
Phosphate	0.01	0.05	0.1	0.75	1.2	0.1	0.05
Chloride	43.4	49.2	52.4	25.14	63.72	41.28	22
TSS	89.41	102.4	112.73	108.12	129.64	67.94	198
TDS	113.41	107.52	164.58	141.4	152.8	103.12	203.61
Chromium (VI)	0.00	0.00	0.01	0.01	0.01	0.01	0.01
*Iron io*ns	0.01	0.05	0.01	0.05	0.1	0.1	0.5

Indicators----------Sampling Dates----------							
	13/8/2012	11/9/2012	12/10/2012	8/11/2012	6/12/2012	8/1/2013	12/2/2013
Temp.℃	18	17	15.5	16	14.5	15	16
PH	8.4	8.2	7.9	8	8.3	8.1	8.7
Total alk.	13.6	11.8	8.64	12.46	10.48	9.8	16.9
COD $_{(IMn)}$	12.6	36.45	8	17.6	8.45	29.1	8.74
Ammonia ions	0.4	0.35	0.7	0.5	0.3	1.5	2.5
Nitrate ions	8.6	0.2	10	5.7	22	8.8	0.8
Phosphate	0.1	0.1	0.05	0.1	0.12	0.15	0.25
Chloride	48	15	49.63	53.18	47.63	147.9	67.73
TSS	108	46.87	63.8	101.6	212	127.4	446
TDS	132.3	93.41	92.67	195.8	298.6	182.56	220
Chromium (VI)	0.015	0.01	0.01	0.01	0.01	0.01	0.015
Iron ions	0.5	0.01	0.01	0.05	0.5	0.1	0.07

Station 4 of Osumi River corresponds more closely to the center of Berati, due to the geographical position of the river that passes through the city. Values of physic-chemical parameters in this location as pH, Total alkalinity, COD $_{(IMn)}$, Ammonia ions, Nitrate ions, Phosphate, Chloride, TSS, TDS; Chromium (VI) and Iron ions are much lower compared to other stations, especially Station 1 and Station 2, and are close to the values of Station 3.Reduction of pollutants in Station 4 is the result of natural aptitude self-cleaning of rivers

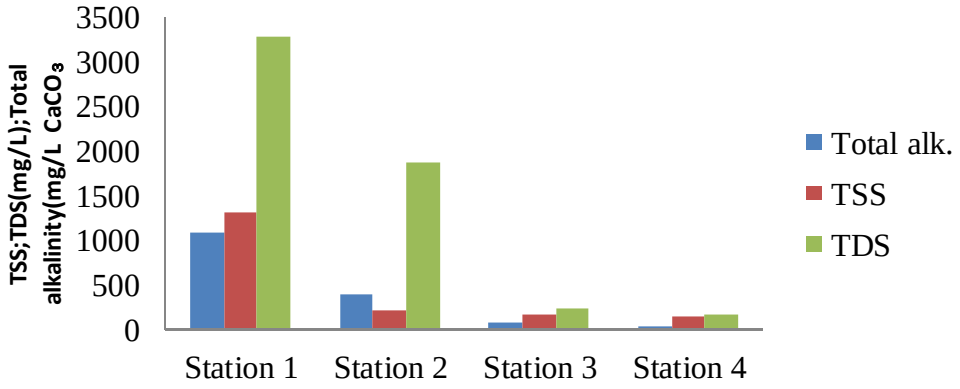

Figure 1 Variation of average values of Total alkalinity, *TSS, TDS* in four stations.

High values of Total alkalinity varies at S1 at limits 89(rainy conditions) - 2245 mg/L $CaCO_3$, at S2 varies from 78.6- 1600 mg/L, at S3 varies 21.4-113.5 mg/L, at S4 5.75-16.9 mg/L. Average value of Total alkalinity at S1 is 1076.99 mg/L, at S2 is 385.72 mg/L, at S3 is 66.15 mg/L, at S4 is 10.168 mg/L. Average values of Total alkalinity at the four

stations differ from 1076.99-10.168 mg/L $CaCO_3$. Values of Total alkalinity depend from pH values. Higher pH, higher alkalinity values, that creates good conditions for the growth of bacterial micro flora. High values of TSS were observed in S1, from 243-2421.6 mg/L, in S2, from 72- 380.52 mg/L, in S3 from 18- 714 mg/L, in S4 from 46.87- 446 mg/L. Mean value of TSS at S1 is 1316.99 mg/L, at S2 are 212.15 mg/L, at S3 is 164.2 mg/L, at S4 is 136.7 mg/L. Higher values of TSS in S1 and S2 are the result of sediments in untreated leather effluents. Values of TSS in S3 and S4 are the result of soil erosion. Higher values of TDS were observed in S1, from 918- 8974.41 mg/L, in S2 from 583.51- 3723.1 mg/L, lower values of TDS in S3 from 61.34- 982.78 mg/L, lower in S4 from 92.67- 298.6 mg/L. Mean value of TDS in S1 is 3283.27 mg/L, in S2 are 1873.55 mg/L, in S3 are 228.88 mg/L, and in S4 are 157.27 mg/L. High values of TDS in S1 and S2 indicate the presence of salts in leather effluents.

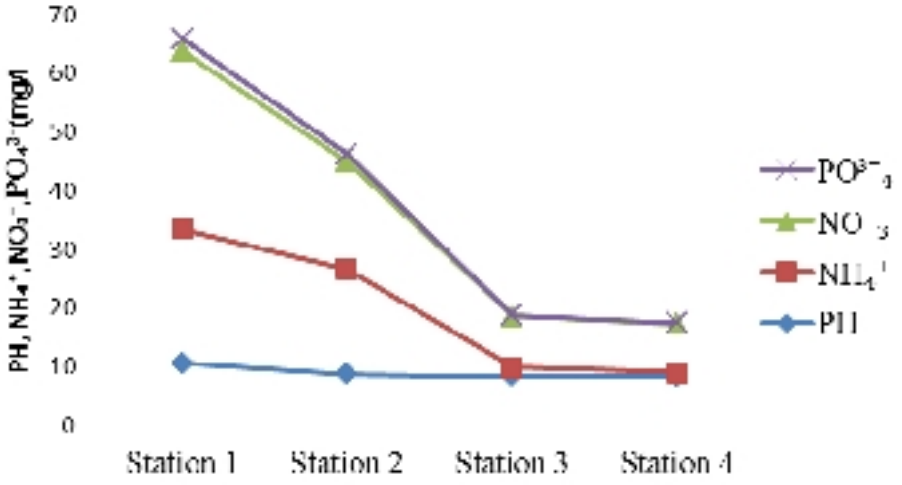

Figure 2 Variation of mean values *of pH, NH_4^+, NO_3^- and $PO4^{3-}$* in four stations

High values of pH varies from 9.3-12.5 in S1 as result of chemicals used in leather processing, in S2 values of pH are 8.2-9.8, in S3 varies from 7.8-8.8, in S4 values of pH varies from 7.8-8.7. Average values of pH result 10.528 in S1, 8.758 in S2, 8.33 in S3 and 8.23 in S4. Values of ammonia ions varies in high levels from 3.52- 45 mg/L in S1, 13-25 mg/L in S2, 0.15-3.5 mg/L in S3, 0.2-2.5 mg/L in S4. Average values of ammonia ions are respectively 22.91 in S1, 17.875 in S2, 1.56 in S3, 0.772 in S4. Higher amount of ammonia ions in S1 is due to decomposition of organic matter in leather wastewater, lower quantity of ammonia ions in S2 compared with S1, is due to dilution of leather effluents in the river basin. Values of ammonia ions in S3 and S4 are due to protein substances produced by flora, fauna and different discharges into river. Values of nutrients as nitrates and phosphates respectively are from 5.7-52.5 mg/L in S1, 10-32.5 mg/L in S2, 0.15-15 mg/L in S3, 0.2-22 mg/L in S4, mean value of nitrates is 30.43 mg/L in S1, 18.375 mg/L in S2, 8.76 mg/L in S3, 8.243 mg/L in S4.Value of phosphate varies from 1.5- 3.25 mg/L in S1, 0.5- 1.25 mg/L in S2, 0.075-0.25 mg/L in S3, 0.01-0.25 mg/L in S4.Mean value is 2.022 mg/L in S1, 1.2 mg/L in S2, 0.127 mg/L in S3, 0.223 mg/L in S4. Higher

values of phosphate in S1 and S2 are due to chemicals used in leather processing, higher value of phosphate in S4 compared with S3 is due to the large urban discharges into river.

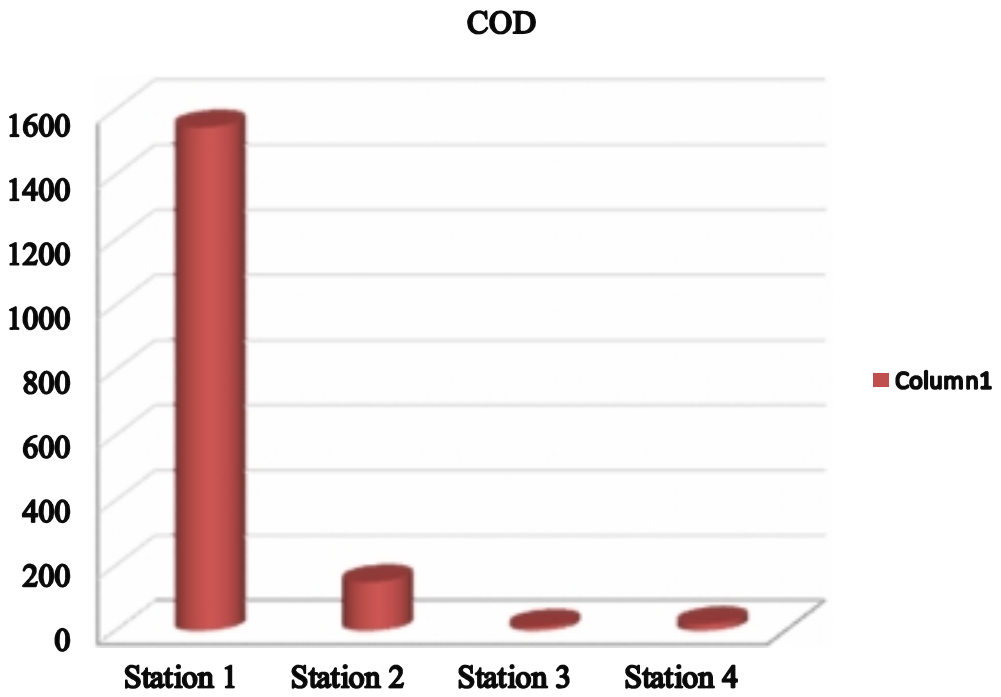

Figure 3 Variation of average values of *COD* $_{(IMn)}$ in four stations

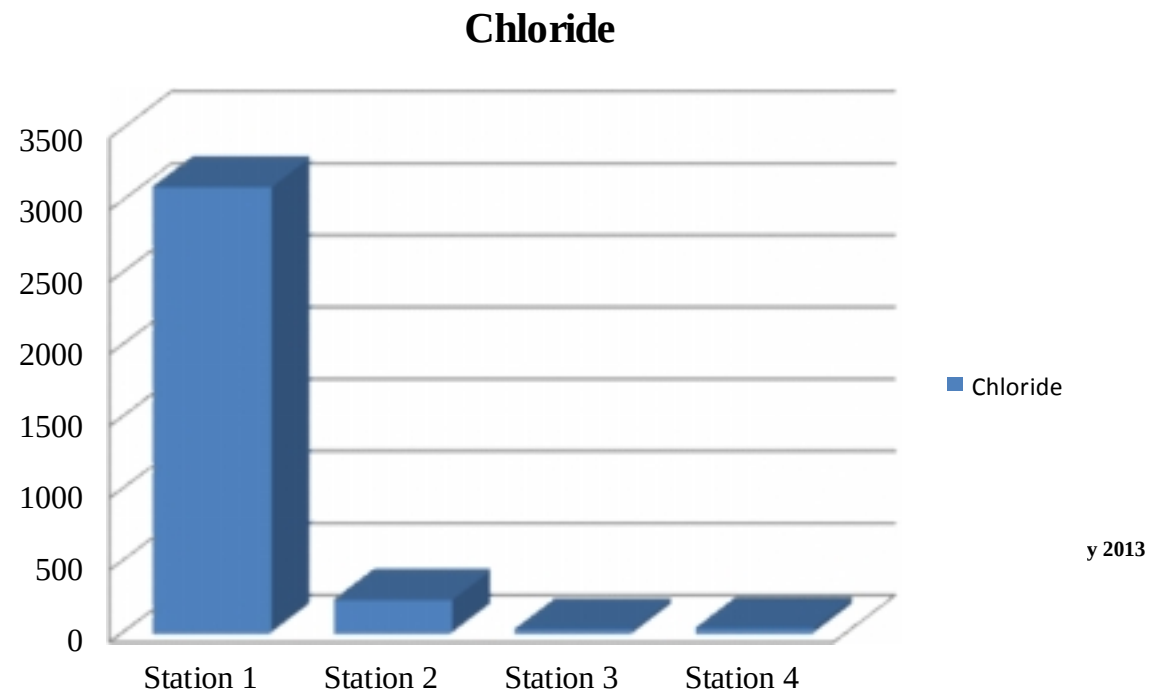

Figure 4 Variation of mean values of chloride *(mg/L)* in four stations

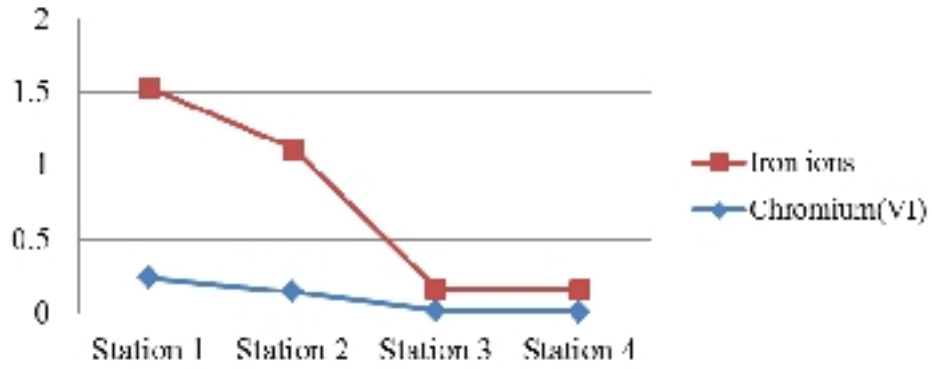

Figure 5 Variation of mean values of Chromium (VI*) mg/L* and Iron ions *(mg/L*) in four stations.

Figure 3 shows the values of $COD_{(I_{Mn})}$ mg/L O_2 resulting from 145.6-11200 in S1, 82.95-272 in S2, 3.2-47.2 in S3, 6.8-38.8 in S4. Mean value of $COD_{(I_{Mn})}$ is 1548.425 mg/L O_2 in S1, 151.91 mg/L O_2 in S2, 14.93 mg/L O_2 in S3, 23.1 mg/L in S4. $COD_{(I_{Mn})}$ express the measure of contamination by organic and oxidizable inorganic matter. High values of COD in S1 and S2 are due to organic leather waste, higher values of $COD_{(I_{Mn})}$ in S4 compared with values of $COD_{(I_{Mn})}$ in S3 are due to large urban waste into river.

Figure 4 shows the values of chloride, respectively vary from 97.725- 10280.5 mg/L in S1, 65.85-850.8 mg/L in S2, 21.25-57.05 mg/L in S3, 15-147.9 mg/L in S4. Mean value of chloride is 3107.14 mg/L in S1, 240.33 mg/L in S2, 37 mg/L in S3, 44.73 mg/L in S4. High value of chloride in S1 and S2 is due to large amount of $NaCl$ used in preservation of raw hides in leather processing. Higher values of chloride in S4 are due to the intrusion of sewage water into river.

Figure 5 shows the values of chromium (VI) mg/L resulting from 0.1- 0.45 mg/L in S1, 0.1- 0.2 mg/L in S2, 0.01-0.03 mg/L in S3, 0.01-0.015 mg/L in S4. Mean value of chromium (VI) in S1 is 0.242 mg/L, 0.145 mg/L in S2, 0.016 mg/L in S3, 0.0108 mg/L in S4. High values of chromium in S1 and S2 are due to the presence of chemical reagents containing chromium in tanning process. Is analyzed the content of chromium in leather effluents, without taking into consideration the amount of chromium deposited in the sediment of the river. In the Figure 5 were recorded and values of iron ions that vary from 0.8-2.5 mg/L in S1, 0.5-1.3 mg/L in S2, 0.05-0.5 mg/L in S3, 0.01-0.5 mg/L in S4. Mean value of iron ions in S1 is 1.29 mg/L, in S2 0.975 mg/L, in S3 is 0.152 mg/L, in S4 is 0.147 mg/L. Higher values of iron ions in S1 and S2 are due to the discharge of leather effluent and sewage into river.

Conclusion

The potential environmental impact of leather industry is significant. Composite untreated wastewater is turbid, colored and smelling. The physical look and smell of area is intolerable, as a result of decaying organic matter in the composition of leather wastewater. The passage of wastewater through the drainage channel that runs along the agricultural lands makes possible contamination of soil layers and ground waters through the infiltration of leather wastewater, particularly chromium. Researchers have reported the infusion of chromium into

plants and ground waters[4]. Osumi River water pollution from leather wastewater is not monitored in this area before. In this sense, this study on its target is an innovation**.**

Suggestions

Further development of leather industry in friendly manner with environment requires the implementation of appropriate techniques for wastewater treatment

Related negative impact of leather effluent on the environment, leather wastewater treatment remains a priority for the near future.

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