Evaluation and Characterization of Wastewater from Tannery industries and their Impact on Water Quality of the Receiving River, Ethiopia

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ABSTRACT

Tannery industries are heavy source of pollution of water body. The objective of this research is to evaluate the current status of physico-chemical properties of tannery wastewater from tannery industries concentrated in Mojo town and their impact on water quality of Mojo River. Accordingly, from seven tanneries, effluents at outlet channel, river water and sediments were collected. Physico- chemical parameters and chromium in samples were analyzed. The results revealed in the ranges of PH (5.85 – 12.04), TDS (3412 - 4660 mg/L), TSS (158.5 -2162.5.5 mg/L), PO_4^{-3} (0.1 – 0.9 mg/L), SO_4^{-2} (456 – 912 mg/L), NH_3 -N (31.5 – 358.2 mg/L), NO_3^{-} (97.4 – 121.3 mg/L), COD (718 -2380.5 mg/L), BOD₅ (706 -2276 mg/L), CI (560.9 - 4779 mg/L), S^{-2} (6.4 – 136.1 mg/L) and Phenol (4.3 – 11.7 mg/L). The study also revealed that the concentrations of chromium ranged from ND - 23.43 mg/L, 0.12 - 1.6 mg/L and 60.1- 264 mg/Kg in effluents, river water and sediments respectively. The mean value of TDS, TSS, SO_4^{-2} , NO_3^{-} CI⁻, ammonia, sulfide, BOD₅, COD in wastewater and river water were above the standard of Ethiopian tannery effluent disposal limits and WHO respectively and Cr in friend ship tannery effluents and river water at site MS02 and LS01, were also above the standard of Ethiopian tannery effluent disposal limits and WHO standard permissible limits. Similarly the level of Cr at the downstream of river sediment was higher as compared to the standard of USEPA sediment quality guidelines. Generally, this study indicated that the river was polluted by direct discharge of tannery wastewater. Therefore, discharging of tannery effluents without treatment is worse for the aquatic environment. The researchers had the inference that, the tannery industries can adopt best available technology and management practice before they discharge their effluents into the receiving water body.

Keywords: Characterization, Tannery, Evaluation, Water quality, pollution.

1. INTRODUCTION

Tannery industry is one of the most environments polluting industrial sector. Almost every tannery industry uses significant amounts of chemicals and water in the process of transforming animal hides into leather [1]. In most developing countries, tannery industries are engaged a wet chrome tanning process that consumes excess amounts of water, and generates about 90% of the used water as an effluents [2]. These effluents contain various organic, inorganic and large quantities of suspended substances [3, 4]. As a result of this, surface water bodies are major receptacles of industrial tannery wastes, have become highly polluted, specially river is main receptor of pollutants [5], because rivers as open systems are more vulnerable to contamination [6], which can alter the physical, chemical and biological nature of the receiving water body [7]. Ultimately, the river sediment also serves as the primary sink for organic and inorganic pollutants [8].

To date, there is a progressive increasing of tannery processing industries in Ethiopia. More than 12 tannery industries are concentrated in Mojo town, Oromia, Ethiopia. About 90% of tannery industries are discharging their wastewater and dumping their chrome solid wastes into open land and nearby water body [9]. Thus, tannery industries are serving as huge source of water pollution by direct discharge of their wastewater without appropriate treatment [10]. Regarding to this, Mojo River is the primary means of receiver of untreated/partially treated tannery effluents (wastewater), disposal of chrome tanning solid wastes. Although, the downstream (lower stream) part of the river is used for livestock production, fish culture, irrigation, and drinking for animals, but it is polluted [11]. The situation is go ahead still now in such away. Therefore, from human health (disappointment of the community around tannery industry due to unstoppable contamination of Mojo River by tannery wastewater) and environmental health point of view the present study was conducted to evaluate the current situation of physico-chemical parameters of tannery wastewater and their impact on water quality of Mojo River so as to generate quality data for concerned body and police makers.

2. MATERIALS AND METHODS

2.1. Study area

The study area is located in Mojo town at 8°35' north latitude and 39°07' east longitude with an elevation between 1788 and 1825 m above sea level, Oromia, Ethiopia. There are more than 12 tannery industries have been concentrated in Mojo Town. However, these industries are discharge their wastewater without treatment, partially treatment through pipe canal directly into Mojo River during night, early morning and dumping their chrome solid wastes near to the river.



Figure 1: Chrome tanning solid waste near to Mojo River (A) and discharge of tannery effluents through canal into the river (B) (Source: Survey and observation)

2.2. Study design

The study involved sampling of wastewater from seven tannery industries, river water and sediment samples at upper stream, near to the point of contact of wastewater with river water(middle stream) and at downstream (lower stream) of the river from the source of pollution were collected.

2.3. Sample collection and Preparation

Tannery wastewater and river water samples were collected according to the standard methods of [12]. The wastewater samples were collected from the outlet of tannery industries: Dx (Pvt) Limited, Kolba (Pvt.) Limited, East Africa (Pvt.) Limited, Hora (Pvt.) Limited, Jian Xin Zhang (Pvt.) Limited, Friend ship (Pvt.) Limited, and Mojo (Pvt.) Limited. In order to show the impact of these tannery wastes on water quality of the Mojo River, water samples were collected above the main discharge of the outlet of the tanneries as control (US03), near to the connection of the outlet effluents of the tanneries to the river water (MS02) and at the downstream (lower stream) of the discharge of outlet effluents of the tanneries (LS01), 200 m far from the site MS02. From each sampling sites, about 500 mL of composite samples were collected using pre-cleaned Polythene plastic bottles.

For the analysis of the concentration of Cr, immediately as the samples to be taken, each sample was treated by 10 ml solution of (2M HNO₃) to protect samples from bacterial attack. Similarly, Composite surface sediment samples were collected by using standard procedure of [13] from the same sampling site of river water having 200g at a depth of 0 to 5cm using a portable auger and stored in clean polyethylene bags. Then, each sample were placed into an icebox to protect them from sun radiations and finally transported to the laboratory for physico-chemical analysis. The collected river water and effluent samples were filtered through filter paper (Whatman 41, Diameter 125 mm) to remove large materials.

The filtrates were used to analyze different physicochemical parameters and air-dried sediment samples were homogenized, ground using a pestle and mortar, then sieved through 63 μ m sieves and stored in a clean polyethylene zip-lock bag and kept refrigerated until further analysis.



Figure 2: Shows map of study area



Figure 3: Sampling sites at outlet of the selected tanneries and Mojo River

2.4. Physico-chemical Analysis of Samples

 P^{H} and TDS was measured in situ using portable P^{H} and TDS meter respectively. Physico-chemical characteristics of samples using parameters viz: Total alkalinity, COD, total suspended solids (TSS), nitrate (NO₃⁻), ammonia (NH₃), phosphate (PO₄⁻³), sulphate (SO₄⁻²), chlorides (Cl⁻), sulfides (S⁻²), biological oxygen demand (BOD5) chemical oxygen demand (COD), phenol and concentration of chromium (Cr) were analyzed as per standard procedures [14, 15]. For extraction and estimation of Cr from sediment samples, mixed acid digestion was used. Briefly, sediment samples of 0.2 g were accurately weighed and placed in a dry and clean beaker and the digestion was carried out with 20 mL of a mixture of Conc. HCl and HNO₃ at a 1:3 ratio (v/v) on a hot plate and the mixture heated to almost dryness, then 20 mL of 0.5M HNO₃ was added and the solution was filtered through Whatman N<u>0</u> 42 filter paper. The filtrate obtained was made up to 100 mL mark with distilled water in volumetric flask and the solution was analyzed for determination of Cr concentration. The results were compared with effluent limit values of the Ethiopian Environmental Protection Authority [16].

Physico-chemical parameters	Methods and instrument		
P ^H	P ^H meter		
TDS	TDS meter		
Total Alkalinity	Titrimetric method		
BOD	Respirometric method, Lovibond meter		
COD	Open reflux method with Back titration		
CI	AgNO ₃ Titrimetric method		
NO ₃ -	Hatch spectrophotometer		
PO ₄ -3	Hatch spectrophotometer		
TSS	Gravimetric method		
SO_4^{-2}	Hatch spectrophotometer		
S ⁻²	Hatch spectrophotometer		
Cr	ICP-OES		

3. DATA ANALYSIS

After physico-chemical analysis, Microsoft Office Excel was used for analysis and presentation of data in comprehensive way. Tables and graphs were used for the presentation of results. All analyzed parameters were then compared with standard of Ethiopian tannery effluents discharge limit, surface water quality standards set by World Health Organization (WHO) and United State Environment Protection Authority (USEPA) sediment quality guide line (SQGs).

4. RESULTS AND DISCUSSION

4.1. Physico-chemical characterization of samples

The physico-chemical parameters of samples collected from the seven tannery wastewater and river water are shown in Table 2 and Table 3 respectively.

Parameters			Tannery in	ndustries					ETED limit
	Dx	Friend ship	Kolba	Hora	Jian	Xin	East Africa	Мојо	Value
					Zhang				
P ^H	8.335	5.85	7.815	7.71	8.425		7.875	12.04	6-9
TDS	3660	3412	3884	4112	3718		4457	4666	1000 mg/L
TSS	263	836	658	1766	246		2162.5	158.5	50 mg/L
TA	3100	2500	300	4700	2600		3900	5300	1000 mg/l
Cl	4779	560.85	3299.45	3087.45	1622.55		2643.95	2010.95	1000 mg/l
NO ₃	103.7	97.4	121.2	112	106		101.5	97.6	50 mg/L
SO_4^{-2}	744	504	912	816	876		754	456	250 mg/L
PO_4^{-3}	0.5	0.1	0.7	0.7	0.3		0.9	0.1	10 mg/L
S^{-2}	36.165	6.4	45.71	38.55	136.05		48.4	14.6	1 mg/L
Ammonia	297.5	31.5	294.8	358.2	243.1		281.4	98.4	30mg/L
BOD	736	1450	1207	2397	902		2576	1133	200 mg/L
COD	716.95	1178.2	1185.8	2380.45	827.95		2420.55	888.4	500 mg/L
Phenol	7.65	4.26	11.64	10.98	8.16		7.38	9.6	1 mg/L
Chromium	ND	23.43	ND	ND	ND		ND	ND	0.1 mg/L

 Table 2: Physicochemical results of tannery wastewater collected from seven tannery industries

ETED- Ethiopian Tannery Effluent Discharge limit, the results are expressed in mg/L except P^H

Table 3: Physico- chemical characteristics of River water, the results are expressed in mg/L

Parameters	US03	MS02	LS01	Standards (WHO, 2008)
H				
P ^H	7.35	9.2	8.91	6.5-9.5
TDS	1537	3393	3230	1000 mg/L
TSS	88	360	128	50 mg/L
TA	771.7	2242	1324	1000 mg/l
Cl	200.9	2940	1360	250 mg/l
NO ₃	17.5	77.7	104.5	50 mg/L
SO_4^{-2}	241.1	548.8	308	250 mg/L
PO_4^{-3}	0.04	0.52	0.013	10 mg/L
S ⁻²	0.375	21.8	8.2	1 mg/L
NH ₃ -N	0.32	96.5	61.6	30 mg/L
BOD	105	954	482	200 mg/L
COD	197	1356	518	500 mg/L
Phenol	0.36	2.74	1.06	1 mg/L
Chromium	0.12	1.6	0.75	0.05mg/l

4.1.1. P^H, TSS and TDS

The P^{H} values in effluents from selected tannery industries ranged from 5.85 to 12.04 (**Table 2**). Highest value (12.04) was observed at Mojo (Pvt.) Limited tannery industry, while its Lowest value of P^{H} (5.85) was observed at Friend ship (Pvt.) Limited tannery industry. The P^{H} value at Mojo (Pvt.) Limited tannery (12.04) was above the standard value of Ethiopian tannery effluent discharged limit into inland water. The alkaline nature in this tannery effluent is due to the presence of lime, soda ash, sodium sulphide and caustic soda used in the hides and skins processing.

The P^{H} of the river water samples was varied from 6.7 to 8.39. The P^{H} values in effluents and river water are within permissible limit of Ethiopian tannery effluent discharge limit and water quality guide line set by WHO, except effluents from Mojo (Pvt.) Limited tannery that is exceeding the limit. TSS and TDS contents of the analyzed tannery effluents were ranged between 158.5-2162.5 mg/l, 3412-4660 mg/l respectively. The highest TSS concentration was recorded in the effluents generated by East African (Pvt.) limited tannery, while lowest concentration was recorded in effluent of Mojo (Pvt.) limited tannery. Higher concentration of TDS (4660 mg/L) was obtained in tannery wastewater discharged from Mojo (Pvt.) limited tannery, this is due to the presence of dissolved inorganic and organic contaminants. In all effluent samples both TSS and TDS concentration were exceeding Ethiopian tannery effluent discharge limit. TSS and TDS value in river water was found in the range of 88- 360 mg/L, 1537 – 3393 mg/L respectively. The maximum TSS and TDS were observed 360, 128 mg/L and 3393, 3230 mg/L at MS02 and LS01 respectively, which were greater than the permissible standards set by WHO (360, 3393 mg/L respectively). Higher values of TDS in these sites are due to the presence of dissolved organic and inorganic substances.

4.1.2. Biological and Chemical Oxygen Demand (BOD₅, COD)

The mean values of BOD₅ in wastewater from the selected tanneries were in the range 1765 -3227.5 mg/L (Table 2). Maximum value of BOD was observed in effluent from East Africa (Pvt.) limited tannery (3227.5 mg/L) and its lowest value observed in effluent generated from Dx (Pvt.) limited tannery (1765mg/L). All BOD₅ values from the tannery effluent obtained were higher than the discharge limits Ethiopian tannery effluent. The high value of BOD₅ at East Africa (Pvt.) limited tannery indicates the presence of massive amount of organic substances present in waste water. Larger concentration of organic constituents utilizes large quantity of O_2 and enhances the level of BOD₅. The depletion of oxygen creates stress on many aquatic organisms including fish. Effluents discharged into aquatic ecosystems alter pH, increase the BOD5 and COD. While, the results in present study also show that BOD values of the analyzed river water samples were varied from 201.6-1458 mg/L. BOD values obtained from river water samples were in the order of : MS02 (1458) > LS01 (523.4) > US3 (201.6). These values at MS02 and LS01 sites were exceeded the permissible BOD₅ level of WHO (200 mg/L), this indicates that the river water is unsuitable for the existence of the aquatic organisms, due to the reduction in the dissolved content.

In the present study, COD values in the tannery wastewater were in the range 716.7to 2420.5 mg/L (Table 2). High level of COD was observed 2420.6 mg/L in effluent discharged by East Africa (Pvt.) limited tannery and lowest value was observed in effluent of Dx (Pvt.) limited tannery (716.7 mg/L). The results also indicated that, COD level at all monitoring tannery industries which did not meet the standard discharge limit into inland surface water (permissible limit:500 mg/L). COD level commonly indicates the concentration of organic matter in wastewater which is not decomposed by microorganisms [17].

The value of COD level in river water was in the range 197- 1356 mg/L. The maximum COD was observed at MS02 site (1356 mg/L), it exceeds the permissible COD level of WHO (500 mg/L) and lowest value was obtained in samples from US03 site (197 mg/L) of Mojo river water, this indicates that the river water is unsuitable for the existence of the aquatic organisms, due to the reduction in the dissolved content.

4.1.3. Alkalinity

In the present study, the alkalinity was ranged from 2500 to 5300 mg/L in tannery wastewater (Table 2). The maximum value of Alkalinity (5300 mg/L) was observed at effluents discharged by Mojo (Pvt.) Limited tannery and Lowest total alkalinity was observed at Friend ship (Pvt.) Limited tannery (450 mg/L) wastewater. The levels of alkalinity at all the sampling sites were higher than the standard permissible limits of Ethiopian tannery effluent discharge limit. The high level of total alkalinity in the sample depends on its high carbonate and bicarbonate concentrations, and indicates the presence of high levels of dissolved salts in the effluents. The total alkalinity in the river water at sites LS01, MS02 and US03 was found to be 1324, 2242 and 771.7 mg/L, respectively (Table 3). The levels of alkalinity at MS02 and LS01 sampling sites were higher than the standard permissible limits set by [18].

4.1.4. Sulphate and Chloride

The mean sulphate and chloride concentrations in wastewater from seven tannery industries were given in Table 2. The results indicate that in the tannery wastewater, the values of sulphate were ranged 456 to 912 mg/L and those of chlorides were ranged 560.85- 4779 mg/L. The results reveal that the effluents of Kolba (Pvt.) Limited tannery contained very high concentration of sulphate than those at the other tannery effluents. This was due to the use of significant amount of sodium sulphide salts. The high concentrations of sulphate in this tannery wastewater may also result from many auxiliary chemicals used. The values sulphate ion in wastewater from each tannery was high compared to the Ethiopian tannery effluent discharge limit. Concentration of sulphate in river water samples also were ranged from 386 – 1580 mg/L. Highest concentration of SO₄⁻² was observed at MS02 (1580 mg/L) and LS01 (1345 mg/L) and those values were higher than water quality standards set by [18]. Chloride (CI⁻) concentration in the effluents from each tannery industry was found to be higher than the Ethiopian tannery effluent discharge limit (1000 mg/L). Presence of high level of chloride in tannery effluents is the result of addition excess amount of NaCl for the preservation and pickling processes of skins and hides. Thus, it remains as a burden in the environment [19]. The chloride concentration in river water also ranged from 586 – 2132.66 mg/L. The maximum Cl⁻ level was observed 2132.66 and 943.66 mg/L in water sample at MS02 and LS01 respectively and exceeds the permissible limits of [18]. Hence, high value of chlorides in effluents resulted in increasing of chloride contamination of river water.

4.1.5. Phosphate and Sulfide

The results of present study revealed that the levels of phosphate and sulphide in the tannery wastewater were ranged from 0.1 to 0.9 mg/L and 14.6 - 136.1 mg/L respectively (Table 2). The values of phosphate in each wastewater samples were within the discharge limit set by Ethiopian tannery effluent discharge limit which is 10 mg/L, whereas sulphide content in tannery effluents were higher than tannery effluent discharge limit of Ethiopian tannery effluent of discharge into inland surface waters.

The concentration of phosphate and sulphide in river water samples were also ranged from 0.08 - 0.76 and 0.64 - 21.8 mg/L respectively (Table 3). The results also indicate that the level S⁻² in water samples collected at MS02 and LS01 site of Mojo River exceeded the standard limits set by [18]. This indicated that the river is polluted by this of pollutant as the result of the direct discharge of tannery wastewater. Sulfides are particularly objectionable because hydrogen sulfide will be liberated if they are exposed to a low P^H environmental, and if they are discharged into stream containing iron, black precipitates will be formed. Sulfides may be toxic to stream organisms or to organisms employed in biological treatment systems.

4.1.6. Ammonia-N and Nitrate

The concentrations of ammonia-N and nitrate in the tannery wastewater ranged from 31.5 to 358.2 mg/L and 97.4 to 121.3 mg/L respectively (Table 2), and the highest values of ammonia were observed in wastewater collected from Hora (Pvt.) limited tannery (358.2) and **Dx** (Pvt.) limited tannery (297.7mg/L), whereas the highest concentration of nitrate was obtained in effluents of Kolba (Pvt.) limited tannery (121.3 mg/L) and Hora (Pvt.) limited tannery (112 mg/L). All these values were above the Ethiopian tannery effluent discharge limit which is 30 for ammonia and 50 mg/L for nitrate.

These high levels of ammonia-N and nitrate might be attributed to several components in tannery effluent containing nitrogen as part of the chemical structure and the nitrogen contained in proteinaceous material of the skin [19]. The concentrations of ammonia-N and nitrate in river water samples were varied from 99.34 to 55.54 mg/L and 143.50 to 78.13 mg/L respectively (Table 3). The levels of ammonia-N and nitrate in the downstream (lower stream) (MS02) site of the river were above the values

obtained from the upper stream site of the river (US03). Thus, this shows that the downstream site of the river is polluted due to the discharged of tannery wastewater.

4.1.7. Concentration of Chromium (Cr)

Wastewater generated from tannery industries is the major source of Chromium pollution. The chromium (Cr) is well-known to be toxic to living organisms due to their bioaccumulation and non-biodegradable properties. According to the research done somewhere else [22] chrome tanning processes originates toxic metals and regular treatment systems are not eligible for the elimination of it. In the present study, concentration of chromium (Cr) measured at all selected tannery industries varied from ND to 23.43 mg/L and found within permissible limit except Fiend Ship (Pvt.) Limited tannery. Cr concentration in effluents from Fiend Ship (Pvt.) Limited tannery (23.43. mg/L) was the highest from the other tanneries. This concentration of Cr was also found to be higher than the prescribed by Ethiopian tanner effluent discharge limit. The disposal of such type of tannery waste water is not safe to discharge into water bodies without proper treatment because they alter the physico-chemical properties of the water creating harmful effect to aquatic ecosystem and the people residing in tanning industrial area are at high risk facing deleterious health effects even on dilution.

While, the concentration of Cr in the river water samples was ranged 0.12 to 1.6 mg/L (Table 3). The highest value of Cr was obtained at sampling site MS02 (1.6mg/L) and LS01 (0.75 mg/L) as shown in Table 3. The values of Cr at site MS02 were higher than those at the other monitoring points LS01 and US03, as expected, and decreased with increasing distance, indicating sedimentation and coagulation of the ions during drainage. The results also indicate that the concentration of Cr in the collected water samples exceeded the standard set by WHO standard. The concentrations of Cr with respect to sampling locations followed the order: MS02 > LS01 > US03.

The concentration of Cr in sediment samples was ranged from 60.1 to 264 mg/Kg (Table 5) with an average of 184.7 mg/Kg. Highest concentrations of Cr were found near to the source of pollution that is in the sampling site of MS02 and concentration of Cr in sediment samples from site MS02 and LS01 were above the Average shale value (90 mg/Kg) and Sediment quality guidelines (SQGs) set by (USEPA, 1999), which indicate that there is chromium pollution load on surface of sediments in these sampling site that might be come from tannery wastewater and due erosion of chrome tannery solid waste that is deposited near to the river bank in rainy season.

Sampling Site	Concentration of Cr in mg/Kg	Reference
RWS1	60.1	-
RWS2	264	-
RWS3	230	-
Average value	184.7	This study
Average shale value (ASV)	90	[20]
Sediment quality guidelines(SQGs)	25	[21]

Table 4: Cr concentrations (mg/Kg) in surface sediments in Mojo River

5. CONCLUSIONS

The pollution from the effluents of the leather industries in Bangladesh is a major environmental and social concern. The results of the study show extremely high values of TSS, TDS, TA, BOD₅, COD, Cl⁻, S⁻², SO₄²⁻, NH₃-N, Phenol, and Cr in the effluents and river water collected from the seven selected tannery industries and three in river water. The values were far above the standard permissible limits for Ethiopian tannery effluent discharge limit. The results also show that the physico-chemical parameters of river water at three different monitoring sites (LS01, MS02 and US03) exceeded the standard permissible limit set by (WHO, 2008). With respect to the concentrations of Cr, the locations followed the order MS02 > LS01 > US03, indicating a decrease of concentration with distance. The study observes that coagulation and sedimentation occurred during river water runoff from one site to the other. The concentration of Cr in the sediment samples was in the order: MS02 > LS01 > US03. The values were far above the standard permissible limits set by (USEPA, 1999) SQGs, indicated high pollution load of chromium in the surface river sediments, and suggested that the untreated leather industrial effluents were not suitable for discharging into surface water and surrounding fields. Such practices pose threats to humans, aquatic life and the entire environment. Therefore, care

should be taken to check the quality of the tannery effluents at regular intervals and to make their necessary treatments, so that the pollutants may not enter the environment.

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