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-RESEARCH ARTICLE-

Characterization of Wastewaters Obtained from Hatay Tanneries

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Abstract

The leather tanning industry is one of the most significant pollutants in terms of both conventional and toxic parameters. On the other hand, leather industry has an important economic role both in Turkey and in the World. In this study, wastewater samples were taken from 15 different tanneries in the Hatay Region. Wastewaters obtained from liming process and chromium tanning process was analyzed. Sulfide, chromium (III), chromium (VI), oil and grease, total suspended solids (TSS), organic matters, biochemical oxygen demand (BOD), chemical oxygen demand (COD), pH and alkalinity were determined according to Turkish Standard Methods.The determined averages values belong to wastewaters obtained from liming process were as following: pH 11.71; COD 16821 mg L^{-1}; BOD 4357 mg L^{-1}; TSS 39023 mg L^{-1}; oil and grease 364 mg L^{-1}; S^{-2} concentration 802 mg L^{-1}; alkalinity 2115 mg L^{-1}. The determined averages values belong to wastewaters obtained from chromium tanning process were also as following: pH 4.23; COD 6740 mg L^{-1}; BOD 377 mg L^{-1}; Cr^{+3} concentrations 372 mg L^{-1}; Cr^{+6} concentrations 127 mg L^{-1}; TSS 14553 mg L^{-1}; oil and grease 343 mg L^{-1}.The results of all analyzes were higher than wastewater discharge standards. As a result, it's necessary to use more effective treatments in order to reduce the negative impacts of leather tanning industry that affect environment, natural water resources and at last human health and welfare.

Keywords:
Leather tanning industry, wastewater, COD, BOD, chromium, sulfide

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Introduction

In the leather tannery industry, animal hides are transformed into leather in a succession of many complex stages, consuming high quantities of water and using large amount of chemicals. Tannery wastewaters are mainly characterized by several key parameters such as sulfide, chromium, nitrogen, oil and grease, suspended solids, COD and BOD. The beamhouse and tanning processes are the most contaminating: the former mainly because of its high organic load and sulfide content, the latter principally because of inorganic salts of chloride, ammonia, chromium and sulfate (Cooman et al., 2003). For this reason, wastewaters obtained from liming and chromium tanning processes were used in this study.

Hatay is located in the eastern Mediterranean region of Turkey. The province lies the banks of the Orontes (Asi) River, approximately 22 km inland from the Mediterranean. There are about 15 small sized leather tannery enterprises in Hatay Region. These tanneries are located inside of the town and their waste waters are released to the Orontes River without any purification process. The daily quantity of the liming effluent and of the chromium tanning effluent is about 50 m^3, of which 55-65 % is from the liming process and 35-45 % from the tanning process. The polluted waters from the liming process are discharged in to the river separately from the polluted waters of the chromium tanning process. This situation is caused an important risk in natural physical environment.

The technological process can be separated in two parts: liming process and tanning process. In liming process: The skins are first washed to remove dirt and blood. Then they are soaked in special vats of cold water for the removal of salt and for softening. The next step is the removal of muscle and fatty tissue adhering to the corium layer by means of revolving knives. Liming is done to swell the skins for the better penetration of the tanning agents and for the hair removal. The unhairing of the hides is accomplished mechanically by means of rolling knives. The skins are washed and then subjected to a process known as bating. After bating, the hides are ready for tanning. The bated hides are first soaked in a solution of sulfuric acid and salt (5-15 kg / 1000 kg raw hide), for 10-16 hours. This operation is called pickling. After pickling, the hides are kept in contact with the tanning solution containing salts of Cr^{+3} (such as $Cr_2(SO_4)_3$). After tanning, the produced leather is lubricated to make it soft. A series of operations known as finishing processes are carried out to produce different types of leather (Tünay et al., 1995). Manufacturing steps were shown in Figure 1.

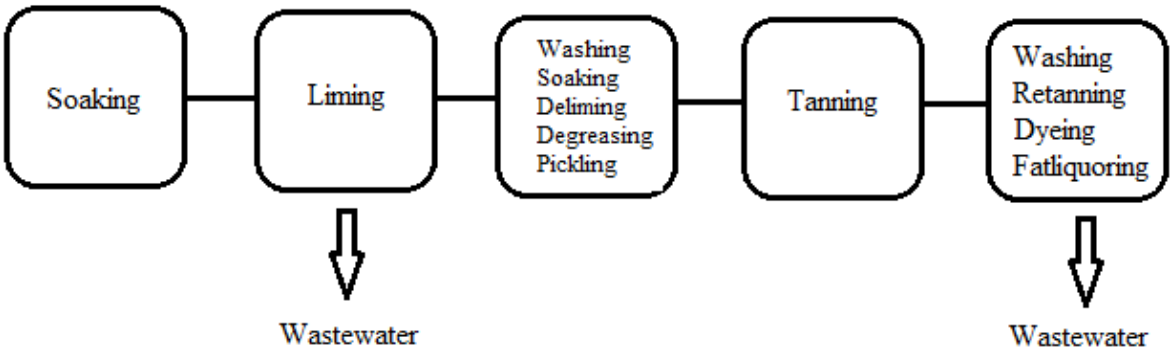

Figure 1. Manufacturing steps.

Wastewater discharge regulations are help to reduce the pollution of waters and maintain the quality of life of the community. Water discharges standards belong to various countries were described in Table 1.

Table 1. Wastewater discharge standards belong to various countries (Saydam, 1998)

Country	BOD	COD	S^{-2}	Cr^{+6}	Total Chromium	Oil and Grease
USA	50	-	0.6	-	1.0	-
England	30	-	0.5	0.1	2.0	-
France	50	125	0.35	0.1	-	-
Germany	-	250	-	-	1.0	-
Italy	40	250	0.8	0.2	2.0	20
Turkey	150	250	2.0	0.5	3.0	30
Argentina	50	200	-	-	0.5	100
Australia	40	-	0.6	-	0.3	-
Austria	30	200	0.3	-	1.0	20
China	150	300	2.0	-	1.5	-
Denmark	-	-	0.3	-	0.2	5
Greece	40	150	0.5	0.5	2.5	20
Hungary	-	150	-	1.0	-	50
Japan	160	160	2.0	-	2.0	30
Holland	5	-	0.8	-	0.05	-
Spain	300	150	2.0	-	1.0	10
Swiss	20	-	0.2	0.1	2.0	20

In the literature, there are a number of studies for the characterization of tannery wastewaters (Szpyrkowicz et al., 1991; Şengül & Gürel, 1993; Tünay et al., 1994; Namasivayam & Senthilkumar, 1994; Ateş et al., 1997; Bajza & Vrcek, 2001; Cooman et al., 2003; Floqi et al., 2007; Mandi et al., 2009; Jahan et al., 2014; Islam et al., 2014). Furthermore, there are also some reviews releated to tanning processes methods and environmental problems (Thanikaivelan et al., 2005; Covington, 1997). But, there is not any study in this matter for the Hatay region of Turkey. The main aim of this study was defined the wastewater characteristics of a group of leather tanning industries located in the Hatay Region.

Materials and Methods

Samples of wastewater were obtained from 15 different commercial tanneries in Hatay Region. Tanneries are in two sections: One is the limehouse which has a separate wastewater channel; the other is the tanyard where pickling, tanning, retanning, fatliquoring and other finishing processes are carried out. This section has also a separate channel. In this study, samples of waste solutions derived from liming and tanning of sheep leather were used. Twelve hour composite samples were taken and stored in 2.5 L polyethylene containers. In the analyses, pH, alkalinity, TSS (total suspended solids), organic matters, BOD (biochemical oxygen demand), COD (chemical oxygen demand), oil and grease and sulfide concentrations were determined according to Turkish Standard Methods. Cr^{+3} and Cr^{+6} and total chromium were determineted by UV–vis spectrometric method, Cr^{+3} is converted to Cr^{+6} using permanganate and 30% H_2O_2. The Cr^{+3} is oxidized to $Cr_2O_7^{-2}$

(dichromate) with $KMnO_4$ and determined coloimetrically using 1,5-diphenylcarbazide to form a purple-violet colored complex. The absorbance of the colored complex was measured in a double beam spectrophotometer, at a wavelength of 540 nm, and chromium concentration of tested samples was determined by a calibration curve prepared using standard solutions of concentrations of Cr^{+6}. All analysis was made in triplicate. All chemicals used were of analytical grade.

Results

Eight main critical parameters belong to wastewaters obtained from liming and chromium tanning processes were shown in Table 2 and 3 respectively. The beam house wastewater was characterized by an alkaline pH (11.71) and the tanning effluent a very acid pH (4.23). The COD values of the liming wastewaters and the tanning wastewaters were found to be 16821 mg L^{-1} and 6740 mg L^{-1}, respectively. The BOD values of the liming wastewaters and the tanning wastewaters were found to be 4357 mg L^{-1} and 377 mg L^{-1}, respectively. The effluents were all characterized by high amounts of TSS (12662 –39023 mg L^{-1}). The high concentrations of S^{-2} (802 mg L^{-1}) and high alkalinity value (2115 mg L^{-1}) were detected in the beam house effluent, whereas the tanning wastewater contained an important amount of total chromium (499 mg L^{-1}). Oil and grease values were found 343 and 364 mg L^{-1} for the liming and tanning wastewaters respectively.

Table 2. Characteristics of the different liming wastewaters

Effluent	pH	COD ($mg.L^{-1}$)	BOD ($mg.L^{-1}$)	TSS ($mg.L^{-1}$)	OM ($mg.L^{-1}$)	S^{-2} ($mg.L^{-1}$)	Alkalinity ($mg.L^{-1}$)	Oil and grease ($mg.L^{-1}$)
F1	11.86	9273	6075	55737	17300	816	2232	240
	(11.80-11.92)	(9200-9350)	(6000-6100)	(55700-55780)	(17000-17500)	(810-820)	(2150-2250)	(235-250)
F2	11.92	6227	6450	78428	18150	768	2328	439
	(11.89-11.96)	(6150-6320)	(6400-6500)	(78400-78460)	(18000-18250)	(760-775)	(2300-2350)	(430-446)
F3	11.80	15650	5750	46540	17200	696	2137	533
	(11.75-11.84)	(1550-15750)	(5700-5760)	(46500-46600)	(17000-17300)	(690-700)	(2100-2160)	(530-540)
F4	11.67	20100	1650	32328	14585	784	1944	311
	(11.70-11.81)	(20000-22000)	(1600-1665)	(32305-32350)	(14500-14600)	(780-785)	(1900-2000)	(305-320)
F5	11.93	10100	2800	24335	13800	960	1968	231
	(11.90-11.96)	(10000-10250)	(2750-2850)	(24320-24350)	(13750-14000)	(955-970)	(1950-2000)	(230-236)
F6	11.24	12600	4750	35200	15300	880	2064	675
	(11.19-11.30)	(12500-12700)	(4700-4800)	(35220-35280)	(15000-15500)	(875-885)	(2000-2100)	(670-680)
F7	11.80	24200	4050	33521	15885	752	1950	423
	(11.75-11.84)	(24100-24300)	(4000-4100)	(35500-35545)	(15750-16000)	(745-760)	(1900-1992)	(410-425)
F8	11.92	25800	5200	37038	15400	1040	2050	308
	(11.88-11.95)	(25700-25850)	(5100-5300)	(37015-37060)	(15300-15500)	(1025-1050)	(2000-2088)	(305-310)
F9	11.93	15200	2600	32435	14650	810	1944	225
	(11.90-11.96)	(15000-15400)	(2550-2650)	(32400-32490)	(14500-14800)	(800-820)	(1900-2000)	(220-240)
F10	11.62	25900	1500	33542	15090	650	1896	276
	(11.59-11.63)	(25750-26000)	(1450-1540)	(33500-33570)	(15000-15150)	(640-656)	(1800-1900)	(275-285)
F11	11.93	15200	2600	32435	14650	810	1944	225
	(11.90-11.96)	(15000-15400)	(2550-2650)	(32440-32490)	(14500-14800)	(800-820)	(1900-2000)	(220-240)
F12	11.62	25900	1500	33542	15090	650	1896	276
	(11.59-11.63)	(25750-26000)	(1450-1540)	(33500-33570)	(15000-15150)	(640-656)	(1800-1900)	(275-285)
F13	11.93	15200	2600	32435	14650	810	1944	225
	(11.90-11.96)	(15000-15400)	(2550-2650)	(32440-32490)	(14500-14800)	(800-820)	(1900-2000)	(220-240)
F14	11.62	25900	1500	33542	15090	650	1896	276
	(11.59-11.63)	(25750-26000)	(1450-1540)	(33500-33570)	(15000-15150)	(640-656)	(1800-1900)	(275-285)
F15	11.93	15200	2600	32435	14650	810	1944	225
	(11.90-11.96)	(15000-15400)	(2550-2650)	(32440-32490)	(14500-14800)	(800-825)	(1940-2000)	(220-235)

Table 3. Characteristics of the different tanning wastewaters

Effluent	pH	COD ($mg.L^{-1}$)	BOD ($mg.L^{-1}$)	TSS ($mg.L^{-1}$)	OM ($mg.L^{-1}$)	Cr^{+3} ($mg.L^{-1}$)	Cr^{+6} ($mg.L^{-1}$)	Oil and grease ($mg.L^{-1}$)
F1	4.10	6200	400	18160	15867	338	104	410
	(4.08-4.13)	(6150-6230)	(390-420)	(18100-18400)	(15840-15890)	(335-340)	(100-110)	(400-415)
F2	4.14	6490	600	13650	2735	346	146	186
	(4.12-4.16)	(6400-6500)	(590-610)	(13600-136700)	(2740-2764)	(340-350)	(140-150)	(180-190)
F3	4.29	9900	310	15300	11058	355	96	406
	(4.28-4.32)	(9750-10000)	(300-320)	(15300-15350)	(11000-11100)	(350-360)	(95-100)	(400-412)
F4	4.26	5300	200	16150	13028	338	158	510
	(4.24-4.28)	(5250-5450)	(190-210)	(16000-16250)	(13010-13075)	(335-345)	(155-160)	(500-515)
F5	4.12	8000	500	12225	7840	359	115	365
	(4.11-4.14)	(7950-8100)	(490-520)	(12000-12500)	(7800-7880)	(355-360)	(110-120)	(360-370)
F6	4.33	5575	300	16240	12045	377	158	153
	(4.30-4.35)	(5450-5600)	(280-310)	(16000-16400)	(12015-12075)	(370-380)	(150-165)	(151-160)
F7	4.19	7575	400	14267	9050	368	85	511
	(4.18-4.20)	(7350-7600)	(390-425)	(14000-14500)	(9000-9100)	(365-370)	(80-87)	(500-523)
F8	4.31	7090	290	15250	10650	360	138	258
	(4.30-4.32)	(6950-7100)	(280-300)	(15000-15600)	(10600-10685)	(355-365)	(135-140)	(253-263)
F9	4.25	8390	200	12300	7835	381	130	436
	(4.23-4.26)	(8250-8400)	(190-210)	(12100-12500)	(7800-7865)	(375-385)	(125-133)	(430-440)
F10	4.04	4300	300	13700	8050	373	151	306
	(4.03-4.06)	(4100-4450)	(280-310)	(13600-13750)	(8015-8100)	(370-375)	(148-160)	(300-310)
F11	4.33	3550	400	11300	7225	342	156	360
	(4.30-4.36)	(3300-3600)	(390-420)	(11000-11400)	(7225-7245)	(340-350)	(150-160)	(355-375)
F12	4.29	6400	450	17250	7427	365	105	278
	(4.28-4.30)	(6450-6500)	(430-470)	(17000-17500)	(7400-7450)	(360-370)	(100-110)	(268-280)
F13	4.12	6850	540	16280	8295	378	116	336
	(4.10-4.14)	(6800-7050)	(525-550)	(16200-16400)	(8240-8340)	(375-380)	(110-120)	(330-340)
F14	4.23	7200	405	12750	10053	383	120	216
	(4.21-4.25)	(7150-7250)	(400-420)	(12500-13000)	(10000-10090)	(380-386)	(110-125)	(200-220)
F15	4.36	7960	355	14200	9743	324	131	410
	(4.34-4.38)	(7950-8100)	(350-380)	(14000-14500)	(9700-9770)	(320-330)	(130-135)	(400-420)

Literature data taken from national and international sources were given in Table 4. As can be seen Table 4, pH, COD, BOD and TSS amounts were within the interval given in the literature. Cr^{+6} concentration and alkalinity values were higher than that of literature values. On the other hand, Cr^{+3} and total chromium concentrations were lower than the literature values. However, the results of all the analysis were higher than wastewater discharge standards.

Many conventional processes are being carried out to treat wastewater from tannery industry such as biological process, oxidation process and chemical process. Among these, physical and chemical methods are quite expensive in terms of energy and reagents consumption. Whereas the mixing method of liming and chrome tanning wastewaters is an extremely convenient method of reducing pollution. Data belong to wastewaters obtained by the mixing of waste solutions derived from liming and chrome tanning were represented in Table 5. As can be seen in Table 5, pH value was reduced to 7.34 in the ratio 1:9 (v/v). TSS, BOD and COD values were reduced from 78428 to 18430 mg L^{-1} and from 6450 to 1300 mg L^{-1} and from 6227 to 3500 mg L^{-1} in the ratio 1:9 (v/v) respectively. The lowest concentration of Cr^{+3} was found in the ratio 9:1(v/v) (98 mg L^{-1}). The smallest value for S^{-2} concentration was observed in the ratio 1:9 (v/v) (200 mg L^{-1}).

Table 4. Pollution Characteristics

Location	pH	COD (mg.L^{-1})	BOD (mg.L^{-1})	Cr^{+3} (mg.L^{-1})	Cr^{+6} (mg.L^{-1})	Total Cr (mg.L^{-1})	TSS (mg.L^{-1})	S^{-2} (mg.L^{-1})	Alkalinity (mg.L^{-1})	Oil and grease (mg.L^{-1})
Antakya*	4.23[a]	6740[a]	377[a]	372[a]	127[a]	499[a]	12662[a]	-	-	343[a]
	11.71[b]	16821[b]	4357[b]	-	-	-	39023[b]	802[b]	2115[b]	364[b]
Niğde[1]	7.30-	1200[c]	-	450-	0.4[c] -	-	1500-	2.5-	600-	50[c]
	8.40[c]	-	-	750[c]	-	-	9000[c]	3.0[c]	1900[c]	-
İstanbul[2]	5.40[a]	1380[a]	-	-	-	1990[a]	-	-	-	-
	12.20[b]	12425[b]	-	-	-	-	-	460[b]	-	-
İzmir[3]	3.11-	470-	3200-	21-	-	-	300-	1.9-	-	40-
	12.51[c]	53800[c]	22500[c]	3372[c]	-	-	73600[c]	5500[c]	-	5400[c]
Çorlu[4]	6.41-	2513-	-	-	-	84-	1000-	10-	259-	-
	10.10[c]	8781[c]	-	-	-	236[c]	4740[c]	121[c]	1132[c]	-
Biga[4]	7.35-	1320-	-	-	-	45-	1365-	26-	345-	-
	9.74[c]	2950[c]	-	-	-	92[c]	2975[c]	82[c]	725[c]	-
Croatia[5]	3.90[a]	6400[a]	2500[a]	561[a]	-	-	-	-	-	-
	12.50[b]	22510[b]	7030[b]	-	-	-	-	8020[b]	-	-
Albania[6]	9.30-	237-	832-	4.75-	-	-	1264-	21-	-	-
	13.00[c]	11032[c]	1631[c]	49.2[c]	-	-	9984[c]	380[c]	-	-
Chile[7]	3.70[a]	7150[a]	-	-	-	-	8600[a]	-	-	-
	12.30[b]	12650[b]	-	-	-	-	87100[b]	2150[b]	-	-
Bangladesh[8]	8.30[a]	12840[a]	4464[a]	-	-	10.348[a]	1250[a]	-	-	-
India[9]	3.34[c]	1912[c]	-	-	-	2375[c]	67403[c]	-	-	-
Italy[10]	7.20[c]	2766[c]	1040[c]	32[c]	-	-	937[c]	56[c]	-	-
Morocco[11]	3.08[a]	2500[a]	45[a]	-	-	1230.5[a]	-	-	-	-
Sudan[12]	3.50[a]	1700[a]	650[a]	-	-	-	46000[a]	-	-	-

[a]Tannery wastewater; [b]Beamhouse wastewater; [c]General wastewater

*This study; [1]Saydam, 1998; [2]Tünay et al., 1994; [3]Şengül and Gürel, 1993; [4]Ateş et al., 1997; [5]Bajza and Vrcek, 2001; [6]Floqi et al., 2007;
[7]Cooman et al., 2003; [8]Jahan et al., 2014; [9] Namasiyavam & Senthilkumar, 1994; [10]Szpyrkowıcz et al., 1991;
[11]Mandi et al., 2009; [12]Islam et al., 2014

Table 5. Data belong to Wastewaters Obtained from the Mixing of Waste Solutions

Liming waste solution: Tanning waste solution (v/v)	pH	COD (mg.L^{-1})	BOD (mg.L^{-1})	TSS (mg.L^{-1})	Cr^{+3} (mg.L^{-1})	S^{-2} (mg.L^{-1})
1:9	7.34	3500	1300	18430	133	200
3:7	7.58	4100	2000	26540	123	250
5:5	8.30	3950	2500	41200	110	330
7:3	8.40	4300	3210	56000	105	400
9:1	8.70	4500	3860	68000	98	600

Discussion

It is revealed with this study that all wastewater analysis results are much higher than national and international wastewater discharge standards. For this reason, wastewater of leather industry must not be discharged to any point before treated.

As a results of literature review, it is seen that physical, chemical, biological, advanced methods and combination of these all methods are utilized in order to improve the quality of leather industry wastewaters.

Ateş et al. (1997) reported that they obtained yields of TSS of 73 %, COD of 51 % and Cr removal by 64 % by pre-precipitation method in leather wastewater. In their study, Kabdaşlı and Tünay (1992) performed chemical precipitation process to leather wastewater and removed 73.9 % of COD while Cr is completely removed.

Şengül et al. (1993) also performed chemical precipitation method utilizing $MnSO_4$ catalyst and achieved 72 % S^{-2} removal. Leta et al. (2004) obtained 98 % COD removal efficiency in their study of effluent wastewater treatment by activated sludge process. Abalı et al. (2014) obtained

50 % COD, 55 % TSS, 33 % BOD and 80 % Cr^{+6} adsorption yields of leather wastewater using clinoptilolite and active clinoptilolite respectively. All of these improvements are carried out at very high costs.

However, significant improvements in pH, COD, BOD, TSS and Cr^{+3}, S^{-2} values can be achieved as we applied at a much lower cost by mixing the wastewater resulting from liming and chrome tanning process at different ratios (v/v). This procedure (mixing 1:9 ratio v/v) has resulted in a reduction of 52.8 % in TSS, 70.2 % in BOD, 79.2 % in COD, 64.24 % in Cr^{+3} and 75.1 % in S^{-2} concentrations.

These procedures can be used in order to obtain more acceptable ecological parameters. Thus, the negative impacts of leather tanning industry that affect environment, natural water resources, at last, human health and welfare can be reduced.

As a result, this study will be an example to the studies which related to reduction of pollution of leather tanning industry in different countries.

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