CHARACTERISTICS OF TYPICAL POLLUTANTS IN TANNERY SITE SOIL: A REVIEW

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Abstract. This paper briefly introduced the process of leather manufacture and the potential pollution sources of soil in tannery sites. Pollutants are mainly derived from the use of a large number of various chemicals and organic matter decomposed by raw skin. The characteristics of typical pollutants in tannery sites soil were summarized, including tannery site soil pH, organic and inorganic compounds, and heavy metals, etc., especially the status of chromium contamination were reviewed. The pH of soil in the tanning workshop (6.65-7.8) is generally lower than tannery sludge dumping site (7.94-8.40). The main organic pollutants contained in the tannery site soil include nitrogen compound, grease, petroleum hydrocarbon. In tannery sludge dumping site soil, the content of nitrogen compound (10cm depth) is 28400 mg/kg, which is similar to tannery sludge. The content of petroleum hydrocarbon is 5-700 mg/kg, which partially exceed the limits of China agricultural land quality standard(<500 mg/kg). In tanning workshop soil, the content of grease is 220-62000 mg/kg. The main inorganic pollutants contained in the tannery site soil include sulfide, high concentration of salt, lime. The high salt content of tannery sludge (99000 mg/kg) leads to high salt content in soil (5500-17500 mg/kg). Total hardness (>450mg/L), total dissolved solids (>1000mg/L), sulfate ions (>250mg/L), nitrite nitrogen (>0.02mg/L) partially exceed the limits of China groundwater quality standard, which are found in groundwater below the tannery site. Heavy metal pollutants in the tannery sites soil have many characteristics and large differences in content, due to the different tanning processes. Among them, chromium (Cr) is the most used heavy metal and the highest content of pollutants. Cr content in tanning process wastewater, dyeing process wastewater and chromium-containing sludge are about 2000-3000 mg/L, 30-40 mg/L and 8500-25800 mg/kg, respectively. Total Cr content in the partial tannery sites soil are higher than 800 mg/kg, which exceed the limits of China agricultural land quality standard(<150mg/kg). Surprisingly, Cr(VI) appears in tannery sites soil and the contents are partly higher than 40 mg/kg, which exceed the limits of China development land quality standard(<3.0mg/kg). Furthermore, the more effort needs to be directed toward the chemistry of chromium-organic complex pollutants, and an understanding of the speciation of Cr in highly organics contaminated tannery site soil is essential for the development of suitable remediation strategies for contaminated soil.

1 Introduction

With the adjustment of industrial structure, China has developed into a world-recognized tanning country. The light leather production reached 735 million m^2 in 2016[1]. The issuance of the "Industrial Structure Adjustment Guidance Catalogue (2011)" promoted the formation of a leather industry cluster area in which the upstream, middle and lower reaches products are mutually compatible. At the same time, some small and medium-sized tanneries with lower concentration are gradually shutting down. For example, Haining City dismantled the old factory building by 16,000 m^2, built and renovated factory building 49,900 m^2 and reclaimed 132,266 m^2 of tannery land. Xinji City established 1.73 million m^2 of tanning industrial zone and bringing together 106 companies[2, 3]. Land used by the tannery may be re-planned as commercial and residential land in the process of industrial restructuring. As the consequence, the remediation and safety assessment before reuse makes the soil pollution problem of the tannery gradually attract people's attention[4, 5]. This paper reviews the pollution characteristics of soils in tannery sites around the world., and puts forward relevant suggestions.

2 Source of Soil Pollution in Tannery

Leather processing technology can be roughly divided into preparation section, tanning section and dyeing and finishing section, and each section contains a plurality of processes. Leather processing has the characteristics of many processes steps and using a large amount of chemical raw materials. Among them, which attracts widespread attention is chromium. More than 80% of the light leather is made from basic chromium sulphate. Wastewater and solid waste are produced in the leather production process. The tannery sludge will be produced after the treatment of wastewater in each section and comprehensive tannery wastewater. According to the statistics, processing 1t cowhide raw material will need 700 ~ 720 kg of various chemical raw materials, produce 60 ~ 120 t wastewater, 150 kg sludge (water mass fraction 70%) and 400 kg meat residue, leather shavings, etc[6, 7]. The typical chrome tanning process, as well as the process of producing wastewater, solid waste and sludge, is shown in Figure 1.

Obviously, heavy metals such as chromium coexist with oil, protein, aromatic, aliphatic, inorganic acid, alkali and salt during tanning. The tanning bath is repeatedly changed from alkaline to acidic according to the process, and the pH span is larger. This leads to complex interactions between chemicals and collagen fibers, and causes complex material transformation, absorption and release during the tanning process. The tannery sludge has a high moisture content and is rich in microorganisms and bacteria. Under natural conditions, the sludge composition is extremely unstable, which makes the migration and conversion of chromium and organic matter in the site more complicated. If the tanning bath leaks, or the tannery and waste shavings percolate during the stacking process, it will lead to complex pollution characteristics of the soil in the tannery.

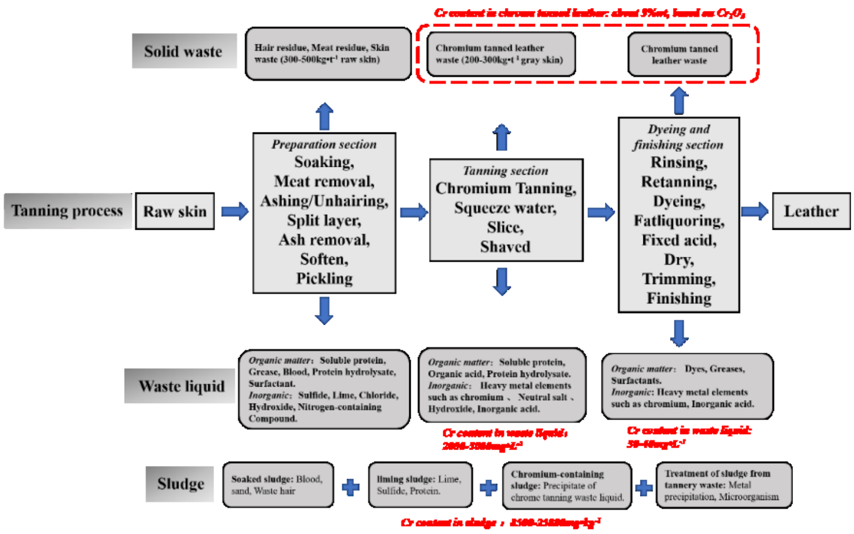

Fig. 1. Major wastes from cattle leather production[8-12].

3 Acidity and Alkalinity (pH) Characteristics of Contaminated Soil in Tannery

pH value is the main physical and chemical index of soil[13]. It not only affects the soil nutrient availability and soil fertility, but also is one of the main factors affecting the environmental effects of pollutants. The survey shows that the soil pH varies greatly in different areas and different depths of the tannery site. This is related to the distribution of tanning workshop, chemical reagents and solid waste dumping site. For example, the pH value of the site soil is found to be 4.57 ~ 7.80 according to the survey of an abandoned 81-year-old tannery in California[14]. The surface soil pH of the tanning workshop is significantly lower than that of the deep soil, showing acidic characteristics. The pH value of the deep soil is closer to the background soil (7.56), indicating that the surface soil is more affected by the acid chrome tanning waste liquid. Kong investigated a site with a 1-year history of tannery sludge dumping in Xinji City, and found that the pH of soil at different depths is about 7.94 ~ 8.40[15]. The pH of the topsoil is similar to the pH of the sludge (7.67), while the pH of the deep soil is closer to the background value (8.30). It indicates that the surface soil is affected by the leachate produced by the tanning sludge stacking, so that the pH value is lower than the background value. A survey of the soil pollution of a tanning solid waste dumping site in Haining City found that the surface soil pH of the site is about 7 ~ 8, and significantly higher than the background surface soil pH (4.67)[16]. The investigation of groundwater in the tanning site area has found that the pH value of each area of groundwater was 6.58 ~ 8.04, and not exceed type of I-III Standard for Groundwater Quality in China[17].

In the tannery site, the pH of the main process bath of chrome tanning is quite different, as shown in Table 1. Obviously, the waste liquid and solid waste pH generated by the tanning site vary with the progress of the process. If the bath droplets leak and the waste leachate enter the site soil, it will inevitably lead to a large difference between the soil pH of the site and the background soil, and present the spatial distribution characteristics of pH closely related to the pollution source of the tanning site. For example, the acidic waste liquid produced in the tanning section gives the contaminated soil an acidic character. During the tanning sludge stacking process, a large amount of alkaline leachate is infiltrated into the soil due to the influence of rainfall, so that the pH value of the soil is alkaline[15].

Table 1. Typical chromium tanning process waste liquid pH.[18-20]

Process	Soaking	Ashing/Unhairing	Ash removal	Pickling	Chromium tanning	Finishing	Integrated wastewater
pH	7.5 ~ 8.0	11 ~ 13	7.0 ~ 9.0	2.0 ~ 3.0	2.5 ~ 4.0	3.5 ~ 4.5	7.0 ~ 9.0

4 Characteristics of Typical Pollutants in the Soil of Tannery

The chemical pollution of the soil in the tannery site comes from two aspects. On the one hand, the leather processing requires the addition of chemical materials with a raw skin content of more than 40%. For example, inorganic salts such as sodium chloride, sulfide, lime, and chromium salts, and organic materials such as softeners, retanning agents, dyes and fatliquors. On the other hand, organic matter decomposition products brought by raw material skin during tannery production, Including oils, animal hair, meat, etc. In leather processing, the combination of these chemical materials and leather fibers is physically or chemically combined, which leads to complex waste water and stacked solid waste components, and change with the process.

Mainly concerned with total chromium, hexavalent chromium, chlorides and sulfides in tannery wastewater and solid waste. In addition, it is generally characterized by comprehensive indicators such as COD, suspended solids, etc, as shown in Table 2. These materials or wastes enter the site soil, undergo complex physicochemical effects, and exhibit various pollution characteristics.

Table 2. Contaminant characteristics of wastewater and sludge in the main process of tanning.[9, 21-25]

	Chloride	Sulfide	Total chromium	Grease	Total nitrogen	BOD_5	COD_{Cr}	Suspended matter
Chromium tanning wastewater, $mg\bullet L^{-1}$	5000 ~ 12500	2 ~ 150	2500 ~ 3000	—	130 ~ 160	100 ~ 250	400 ~ 500	
Dyeing and finishing wastewater, $mg\bullet L^{-1}$	500 ~ 1000	—	30 ~ 40	20000 ~ 50000	400 ~ 500	1000 ~ 2000	2500 ~ 7000	600 ~ 1000
Tannery sludge, $mg\bullet kg^{-1}$	5800 ~ 14000	335 ~ 405	8500 ~ 25800	—	700 ~ 1700	—	10000 ~ 30000	—

Note: "—" No relevant values found

4.1 Typical Organic Pollution in Tannery Sites

The amount of organic materials used in the tannery site is large, and there are many organic residues, resulting in a high total amount of soil organic matter in the tannery. The TOC value in the surface soil (10cm) of the tannery sludge dump site is 14.3%, which is similar to the sludge TOC value. The TOC value in deep soil (100cm) is 0.2%, which is similar to the background soil. The total nitrogen concentration in the surface soil is as high as $28400 mg\bullet kg^{-1}$. Due to soil adsorption and microbial degradation, the total nitrogen and ammonia nitrogen content in the soil decreases with increasing depth. When the depth reaches 70cm, the total nitrogen concentration is reduced to $700 mg\bullet kg^{-1}$ [15]. The oil and grease pollution in the soil of the tannery site is also serious. The content of oil and grease in the surface soil (15cm) can reach $62000 mg\bullet kg^{-1}$. Due to the finer bay mud retardation effect, the oil and grease content was reduced to 220 $mg\bullet kg^{-1}$ in soil with a depth of 137.16 cm[14]. Investigation of a tannery sludge dump in Haining revealed that the total petroleum hydrocarbon content in the surface soil is generally higher than that in the deep soil. The content of petroleum in soil is more than $700 mg\bullet kg^{-1}$, which exceeds the Soil Environmental Quality Risk Control Standard for Soil Contamination of Development Land, and Guideline for Risk Assessment of Contaminated Sites[26, 27]. There are many kinds of organic substances used in tannery sites, but there are few studies on the migration of complexes formed by interaction between different organic substances in soil. The chemical and physical interactions between different organic substances are complex and deserve further study.

4.2 Typical Inorganic Pollution in Tannery Sites

The amount of inorganic salt used in the pre-tanning treatment section is relatively large, among which the main environmental hazards are sulfides, high concentrations of sodium chloride and lime. Unlike organic pollution, inorganic salts are highly mobile and may cause soil and groundwater to be contaminated. The investigation found that the salt content in the surface soil (10 cm) of the tannery site was 17500 $mg\bullet kg^{-1}$. The salt content in deep soil (200 cm) is still high, reaching $5000 mg\bullet kg^{-1}$ [15]. Groundwater survey on contaminated areas in tannery sites indicates that the inorganic matter of the groundwater is characterized by total hardness (256 ~ 606 $mg\cdot L^{-1}$), solubility total solid (456 ~ 1116 $mg\cdot L^{-1}$), sulfate (33 ~ 344 $mg\cdot L^{-1}$), chloride (50 ~ 243 $mg\cdot L^{-1}$), nitrate nitrogen (0.25 ~ 15.02 $mg\cdot L^{-1}$), nitrite nitrogen (0 ~ 0.203 $mg\cdot L^{-1}$) and ammonia nitrogen (0 ~ 0.031 $mg\cdot L^{-1}$)[17]. Inorganic concentration at some sampling points exceed the national groundwater quality standards to varying degrees, and the polluted area is within $0.2 km^2$ of the tannery. Correlation analysis showed that the total hardness of groundwater increased, accompanied by an increase in the concentration of chloride, sulfate, and dissolved solids.

4.3 Typical Heavy metal Pollution in Tannery Sites

There are three sources of heavy metals in the tannery. First, the use of metal chemical materials in the tanning process, including inorganic tanning agent such as chromium, antibacterial agents for heavy metals such as silver, metal complex dyes such as cobalt, inorganic pigments such as cadmium sulfide, Metal flocculant such as polyaluminum chloride[28-32]. The second is the entrainment of the chemical production process, such as organic retanning agents, dyes, pigment pastes[33, 34]. Third, the heavy metals produced by the corrosion of production equipment. Heavy metal pollution in tannery sites varies by process. There are many types and large differences in content, but chromium pollution is the most prominent. For example, in the tannery sludge, Cr content is 11784～20490 $mg \cdot kg^{-1}$、Zn content is 380～864 $mg \cdot kg^{-1}$、Cu content is 70～179 $mg \cdot kg^{-1}$、Ni content is 23～23.82 $mg \cdot kg^{-1}$、Pb content is 13～280 $mg \cdot kg^{-1}$、Cd content is 0.24～3 $mg \cdot kg^{-1}$ [35].

Deepali has found that the concentration of heavy metal elements in the soil of a tannery site are Cr(630.85～815.25 $mg \cdot kg^{-1}$)、Fe(37.45～37.95 $mg \cdot kg^{-1}$)、Mn(0.96～0.99 $mg \cdot kg^{-1}$)、Cu(0.03～0.05 $mg \cdot kg^{-1}$) and Cd(0.03～0.05 $mg \cdot kg^{-1}$), respectively[36]. Cr contamination in groundwater samples (0.93mg/L) was observed only in samples collected from nearby areas of tannery. The findings also indicate that the Cr contamination was more than other metals. Rahaman has investigated the contents of Cr, Pb, Cd and Zn in the soil of the tannery site[37]. The content of various heavy metals in each soil layer is quite different, and the Cr content is 31～561 $mg \cdot kg^{-1}$, Zn content is 73～158 $mg \cdot kg^{-1}$, Pb content is 24～70 $mg \cdot kg^{-1}$, Cd content is 0.71～2.25 $mg \cdot kg^{-1}$. It indicates that the concentration of these heavy metals in the soil of the tanning site is significantly higher than that of the background soil. Andy investigated the content of Cr in groundwater near the tanning site. The Cr content measured from some sampling points is 0.008-0.115 $mg \bullet L^{-1}$ [38]. The reason for the occurrence of Cr in groundwater is that the organic matter produced by the tannery site forms chromium-organic complex, which increases the solubility of Cr in water. In addition, formation of chromium organic complex can prevent oxidation of Cr(III) to Cr(VI). Kashem has investigated the soil and plant heavy metal pollution in tannery sites[39]. The concentrations of Mn, Zn, Cu, Ni, Pb and Cd in the soil are 218～577, 73～477, 35～217, 47～112, 20～89 and 0.87～1.8 $mg \bullet kg^{-1}$, respectively. Heavy metal concentration decreases with increasing distance from the tannery. The total concentration of Cd, Cu, Mn, Ni, Pb and Zn in soil is positively correlated with the extractable concentration of DTPA. Araújo has used visible-near-infrared-infrared spectroscopy (VIS-NIR-mid) to study chromium-contaminated soil in tannery sites[40]. It was found that the adsorption of Cr in the soil leads to a significant change in the spectrum, and the Cr content can be calculated by the spectral reflectance in the VIS-NIR-mid spectrum. Another survey found that some soil samples from the tannery site also detected the presence of Cr(VI)[15, 16], and the content (250 $mg \cdot kg^{-1}$) far exceeded the Soil Environmental Quality Risk Control Standard for Soil Contamination of Development Land.

It should be noted that during the long-term aging process of heavy metals in polluted soils, there is almost no change in the total amount, but its bioavailability gradually decreases with the passage of time. Determination of the total amount of heavy metals in the soil is not suitable for evaluating the bioavailability, toxicity and migration of heavy metals[41-44]. Therefore, more attention should be paid to the content of bioavailable heavy metals in the investigation of heavy metal pollution in contaminated soil at tannery sites [45-50].

5 Conclusion and Outlook

High content and various of organic and inorganic salts in contaminated soil at tannery sites, which is closely related to the tanning process. Among them, Chromium is the main pollutant of concern.

Fractionation analysis showed that the content of effective chromium in the soil of the tannery was significantly higher than that in the background area, which significantly increased the mobility and bioavailability of chromium. Although tanneries use only trivalent chromium, higher levels of hexavalent chromium are found in the site soil. Therefore, when investigating the migration and transformation mechanism of chromium in the tanning site contaminated by compound pollution, attention should be paid to the influencing factors of available chromium and hexavalent chromium. Furthermore, it is necessary to clarify the formation mechanism of soil hexavalent chromium in tannery sites to determine the environmental risk composition of the site. It provides a basis for the development of remediation agents suitable for complex contaminated soil in tannery sites, and also provides an ecological reference for the selection and development of chemical materials for cleaning tanning.

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