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RESEARCH ARTICLE

DETERMINATION OF SELECTED HEAVY METALS CONCENTRATIONS (Cr, Fe, Zn, Pb, Cu, Ni, AND Ti) IN LAWDAR'S TANNERY-YEMEN

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ABSTRACT

This study has been carried out at Lawdar's tannery (Aden-Yemen), and aimed to determine the mean concentration of seven selected heavy metals (Cr, Fe, Zn, Cu, Pb, Ni, and Ti). Four samples (three liquid samples and one solid sample), were analyzed by the ICP-OES technique. The results obtained from this study showed overall mean concentrations of the selected heavy metals in the range of (Cr = 0.066 - 2.366 mg/L), (Fe = 2.230 - 6.519 mg/L), (Zn = 4.430 - 42.029 mg/L), (Cu = 0.033 - 3.166 mg/L), (Pb = 0.133 - 0.866 mg/L), (Ni = 0.033 - 0.699 mg/L), and (Ti = 0.266 - 49.49 mg/L), respectively. The mean concentrations of the selected heavy metals in the wastewater were above the recommended limit.

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INTRODUCTION

Tannery waste is generated in huge amounts during the process of tanning by leather industries throughout the world. It has been considered as one of the most polluted industrial wastes that contain high levels of metals which are very toxic to animals, plants, and soil. Tannery wastes are of serious consequence since it has a role in the pollution of freshwater bodies, streams, and land. Tanneries, oil refineries, and metal industries are causing de-pollution of surface and groundwater quality (Raj, 1996). Industrial liquid effluents are one of the principal sources of heavy metal responsible for environmental pollution (Solomon et al., 2015). The wastewater from leather tannery contained high amounts of toxic chemicals, heavy metals, and other undesirable substances. As such, among all the industrial wastes, tannery effluents have drained the highest pollutants (Umar et al., 2017). In the environment, Heavy metals are toxic and resist bio-degradation which are discharging pollutants from industrial wastewater.

Disposal of effluents from the industries has resulted in serious contamination of numerous sites. Elements such as cadmium (Cd), chromium (Cr), copper (Cu), Lead (Pb), zinc (Zn), mercury (Hg), etc. have toxic effects on human health and also non-renewable resources. Contamination of surface water by the presence of heavy metals from the natural ecosystem, as pollutants adsorption onto the soil and then to the aqueous environment, etc., has caused serious problems to human health (Vani, 2012). The heavy metal content of the soil is of major significance in relation to their nutrient status and fertility. Metals such as Zn, Cu, and Se are essential elements for the normal growth of plants and living organisms. However, deficiency or excess of these metals could lead to a number of disorders. Non-essential and/or toxic elements such as Ni, Pb or Cr, may be allowed by the environment in low concentrations, become toxic in higher concentrations (Yargholi, 2008). On the contrary with the previous study (Saeed, 2021), the recent work focused on analysis heavy metals levels (i.e. chromium (Cr), iron (Fe), zinc (Zn), lead (Pb), copper (Cu), nickel (Ni), and titanium (Ti)) in Lawdar's

tannery effluents to check the effect of the hazard on health and environment by comparing the results with some limits standards.

MATERIALS AND METHODS

Description of the study area: The study was conducted on Al-Harib's tannery, Lawdar town, Abyan Governorate. The tannery is located in the heart of the town near Lawdar Old Fuel Station, on the main road leading to Lawdar General Hospital. It is located about 180 Km to the east of Aden, south of Yemen. The tannery area is about 11,000 m² and consists of three rooms. The first room is used to gather skins from the market. The second room is for chemical processing such as inorganic tanning etc. The third room is for storing tanned leather to export to local and international markets. After tanning, the waste is disposed to sewage.

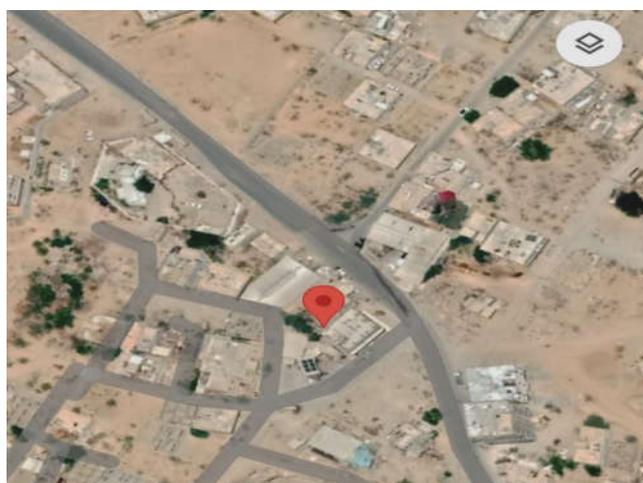


Fig. 1. Map of study location

Samples Collection: The wastewater samples of the tannery were collected during the period from October 2019 to June 2021. Four samples were collected. The first two samples were collected from the tank that was devoted to the wastewater of the tannery. The third sample was a mixed sample of the wastewater from the tank and the soil near the sewage in the study area. The fourth sample was taken from the soil near the sewage to the sewers in the study area. These samples were saved in dark polythene bottles and treated as mentioned in (Jeffery, 1996). The investigated heavy metals were determined by the Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES) technique.

Samples Digestion: After collecting the fluid samples, the digestion started in which 50 mL of the sample plus 5mL of concentrated HNO₃ was heated ~4hr in a high-heat resistant beaker using an electrical heater in order to make yellow sediment.

The process was repeated once again and then the sediment was left to get cold then, diluted by redistilled water (~0.1 μS/cm) up to 50mL solution and became ready to test. For preparing the solid sample, we took one gram from dry soil which was obtained from the soil near of sewage basin in the town by added 15mL of HCl and HNO₃ at ratio 1:3 which was heated on an electrical heater for 4 hr on a thermal plate to make sediment. At that point, redistilled water was added up to 50mL and the solution became ready to test.

Sample Investigation: The analysis process was conducted in the Supreme Board of Drugs and Medical Appliances- Aden, by taking 3mL of digested sample separately and diluting to 100mL. Then, diluted samples were filtered so that the ICP-OES was able to test them accurately.

Reagents and Equipment: All the reagents used were of analytical grade without any further purification.

Statistical Analysis: Statistical analysis was performed using SPSS Statistical Package v20. Analysis was repeated at least three times and data were expressed as mean, standard deviation (±SD), relative standard deviation (RSD%), standard error (±SR), lower bound-upper bound, minimum-maximum (Min-Max) values. and analysis of variance (ANOVA).

RESULTS AND DISCUSSION

Mean concentrations of the selected heavy metals in the tannery effluent are presented in Table (1) and Figure (2).

Table 1. Mean concentration of the selected heavy metals analyzed in the samples, (n = 4)

| Sample No. | Metal | Parameter | | |
|------------|-------|------------------|-------|----------------------------|
| | | Mean ± SD (mg/L) | RSD % | Selected wavelength λ (nm) |
| 1 | Cr | 0.166 ± 0.0001 | 2.23 | 267.716 |
| | Fe | 4.660 ± 0.0007 | 0.53 | 238.204 |
| | Zn | 12.166 ± 0.0019 | 0.52 | 206.200 |
| | Cu | 0.0990 ± 0.0001 | 3.78 | 327.393 |
| | Pb | 0.2000 ± 0.0019 | 32.62 | 220.353 |
| | Ni | 0.0999 ± 0.0004 | 16.83 | 231.604 |
| | Ti | 0.9665 ± 0.0024 | 8.26 | 190.801 |
| 2 | Cr | 0.066 ± 0.00002 | 8.59 | 267.716 |
| | Fe | 2.230 ± 0.0002 | 0.26 | 238.204 |
| | Zn | 4.430 ± 0.0007 | 0.51 | 206.200 |
| | Cu | 0.033 ± 0.0003 | 16.81 | 327.393 |
| | Pb | 0.133 ± 0.0010 | 27.14 | 220.353 |
| | Ni | 0.033 ± 22.31 | 22.31 | 231.604 |
| | Ti | 0.266 ± 0.0021 | 26.08 | 190.801 |
| 3 | Cr | 0.166 ± 0.0002 | 2.98 | 267.716 |
| | Fe | 5.799 ± 0.0011 | 0.61 | 238.204 |
| | Zn | 8.760 ± 0.0034 | 1.28 | 206.200 |
| | Cu | 0.066 ± 0.0001 | 3.51 | 327.393 |
| | Pb | 0.499 ± 0.0004 | 2.50 | 220.802 |
| | Ni | 0.066 ± 0.0003 | 19.43 | 231.604 |
| | Ti | 0.766 ± 0.0024 | 10.07 | 190.801 |
| 4 | Cr | 2.366 ± 0.0005 | 0.68 | 267.716 |
| | Fe | 6.519 ± 0.329 | 1.68 | 238.204 |
| | Zn | 42.029 ± 0.0114 | 0.90 | 206.200 |
| | Cu | 3.166 ± 0.0012 | 1.28 | 327.393 |
| | Pb | 0.866 ± 0.0011 | 4.17 | 220.353 |
| | Ni | 0.699 ± 0.0002 | 0.94 | 231.604 |
| | Ti | 49.49 ± 0.0084 | 0.56 | 190.801 |

The mean concentrations of Cr, Fe, Zn, Cu, Pb, Ni, and Ti in the first sample were 0.166, 4.660, 12.166, 0.099, 0.200, 0.100 and 0.967 mg/L, respectively. The mean concentrations of the same elements in sample 2, were 0.066, 2.230, 4.430, 0.033, 0.133, 0.033, and 0.266 mg/L, respectively. For the third sample, the heavy metals had the mean concentrations Cr (0.166mg/L), Fe (5.799mg/L), Zn (8.760mg/L), Cu (0.066mg/L), Pb (0.499mg/L), Ni (0.066mg/L), and Ti (0.766 mg/L). Finally, the mean concentrations of Cr, Fe, Zn, Cu, Pb, Ni, and Ti in the fourth sample were found to be 2.366, 6.519, 42.029, 3.166, 0.866, 0.699, and 49.490 mg/L, respectively. Heavy metals levels in the analyzed samples (1, 2, and 3) were followed the order Zn > Fe > Ti > Pb > Cr > Cu = Ni, and sample 2 showed the lowest values.

Table 2. The overall mean concentrations of the selected heavy metals in all samples

| Metals* | Mean \pm SD* | \pm SR | 95% Confidence Interval for Mean | | Minimum | Maximum |
|---------|-----------------------|----------|----------------------------------|-------------|---------|---------|
| | | | Lower Bound | Upper Bound | | |
| Cr | 0.6910 \pm 1.1177 | 0.5588 | - 1.0874 | 2.4694 | 0.066 | 2.366 |
| Fe | 4.8270 \pm 1.8931 | 0.9466 | 1.8147 | 7.8393 | 2.230 | 6.519 |
| Zn | 16.8495 \pm 17.0847 | 8.5423 | - 10.3360 | 44.0350 | 4.430 | 42.029 |
| Cu | 0.8467 \pm 1.5466 | 0.7733 | - 1.6142 | 3.3077 | 0.033 | 3.166 |
| Pb | 0.4248 \pm 0.3347 | 0.1673 | - 0.1078 | 0.9573 | 0.133 | 0.866 |
| Ni | 0.2275 \pm 0.324 | 0.1618 | - 0.2872 | 0.7422 | 0.033 | 0.699 |
| Ti | 12.8770 \pm 24.424 | 12.212 | - 25.9867 | 51.7407 | 0.266 | 49.49 |

* Recommended limits (mg/L) by WHO (2006) (8): Cr (0.10), Fe (0.30), Zn (0.03), Cu (0.01), Pb (0.05), Ni (0.02), Ti (---).
By USEPA (2010) (9): Cr (0.10), Fe (---), Zn (2.00), Cu (1.00), Pb (0.015), Ni (0.2), Ti (---).

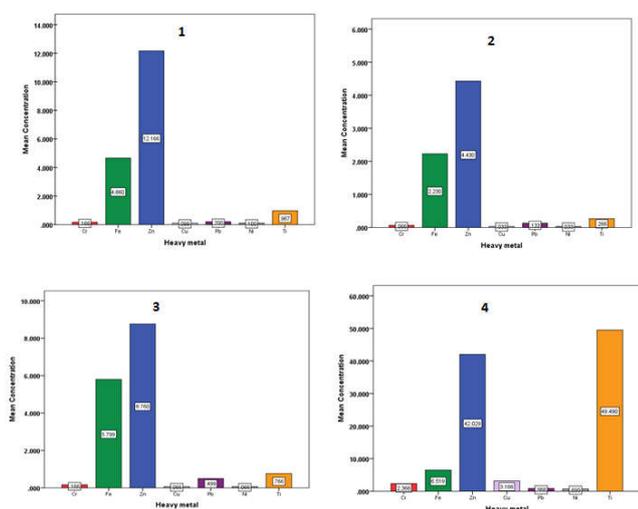


Fig. 2. Mean concentration of the selected heavy metals in samples (1-4)

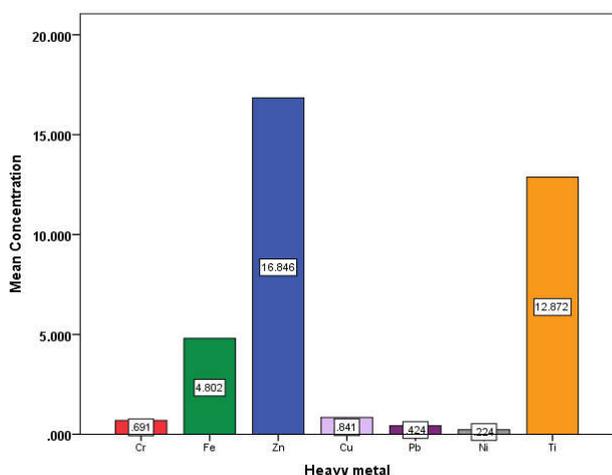


Fig. 3. The overall mean concentrations of the selected heavy metals in all samples

On the other hand, the highest mean concentrations of heavy metals were coined to sample 4 which varied from the above-mentioned samples and followed the pattern $Ti > Zn > Fe > Cu > Cr > Pb > Ni$. In all samples, the mean concentrations of Cr ranged from 0.066 mg/L to 2.366 mg/L. For Fe, the mean concentrations fall between 2.230 and 6.519 mg/L. However, the mean concentrations of Zn ranged from 4.430 to 42.029 mg/L. Cu mean concentration ranged from 0.033 to 3.166 mg/L while Pb mean concentrations ranged from 0.133 to 0.866 mg/L. Ni mean concentrations (0.033-0.699 mg/L) while Ti mean concentration ranged from 0.266 to 49.49 mg/L. The overall mean concentrations of the selected heavy metals in all samples were found to be in the sequence $Zn > Ti > Fe > Cu > Cr > Pb > Ni$. (see Table 2 and Figure 3).

Bernard and Ogunleye (Bernard, 2015) found that the ranges of mean concentrations for Cr, Pb, Fe, and Cu were 3.33 - 5.79 mg/L, 0.67 - 3.10 mg/L, 3.53 - 8.12 mg/L. and 0.82 - 1.51 mg/L respectively which were higher than the maximum permissible limits of WHO and USEPA. Bahiru in his work (Bahiru *et al.*, 2019) concluded that the mean concentrations of Cr in different samples were 0.2 ± 0.00 - 1.04 ± 0.00 mg/L. For Pb, the concentrations ranged between 1.98 ± 0.04 and 3.11 ± 0.04 , mg/L, Fe was between 2.89 ± 0.04 and 5.13 ± 0.06 mg/L, and for Cu found to be between 0.3 ± 0.00 and 0.99 ± 0.06 mg/L. These values are above the permissible limit set by WHO (2006). One study (12) summarized the order of metal mean concentration as $Cr (2.007-3.73 \text{ mg/L}) > Mn (1.421-3.049 \text{ mg/L}) > Zn (0.105-0.934 \text{ mg/L}) > Cu (0.024-1.906 \text{ mg/L}) > Cd (0.015 \text{ to } 0.050 \text{ mg/L})$. The recent work showed that the overall mean concentrations of the selected heavy metals were above the permissible limits, given by WHO and almost USEPA, in all samples. This agreed with Akan with his coworkers (Akan *et al.*, 2019) who found that the mean concentration of metals ions in tannery effluents was very high compared to standards limits. The high concentrations of Zn, Ti, and Fe (Figure 3) in the considered samples may be related to simplicity and cost-effectiveness in using their inorganic salts as tanning agents, sustainable wet-white tanning processes, and the pickling natural leather (Nishad *et al.*, 2013; Kleban, 2006; Ferrer, 2012; Crudu, 2014).

CONCLUSION

The results of this study showed that the mean concentration of the selected heavy metals of the soil sample was higher than that found concerning the fluid samples. The mean concentrations of the select heavy metals in all samples were above the limits set by WHO and USEPA.

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