RESEARCH ARTICLE

Chromium Speciation and Agricultural Soil Contamination in the Surrounding Tannery Regions of Walajaphet, Vellore District, Southern India

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ABSTRACT

The emerging environmental problem in the tanning industry is the disposal of chromium-contaminated sludge in the soil. In the Walajapet Taluk of the Vellore District, the distribution and mobility of chromium in the soils and sludge surrounding the tannery waste disposal region were explored. This paper presents soil contamination of agricultural lands located in the SIPCOT Industrial complex near Walajapet in Vellore District, India. 64 soil samples were collected from 15 different villages, and the soil samples were analyzed for pH, EC, total chromium, hexavalent, and trivalent chromium. Studies were carried out to find the chromium contamination of the agricultural lands due to industrial effluents. It is found that 75 % of the agricultural soil samples were highly contaminated, posing a risk to agricultural lands, based on the comparison of chromium in the soil with WHO standards. The majority of the soil samples (>80%) from the locations exhibited high Cr (>200 mg kg^{-1}) concentrations that exceeded the maximum permissible limit. Furthermore, soil samples taken near tanneries revealed that tannery effluent has a significant impact. The facts strongly demonstrate the existence of hazardous chromium, resulting in severe deterioration of agricultural land. As a result, developing an effective plan and implementing a suitable remediation technique to address the heavy metals contamination problem is critical.

Keywords: *Chromium contamination; Tannery effluent; Chromium speciation; Agricultural land; Remediation*

INTRODUCTION

Population explosion has resulted in resource overexploitation and unregulated industrial waste dumping in the environment. For instance, the growth of tannery industries has resulted in chromium contamination leading to varying degrees of severity and pollution levels (Ferronato and Torretta, 2019). In addition to As, Cd, Hg, Pb Cr (VI) is also a toxic heavy metal at very low concentration; in humans, they cause severe health injury and impairments in the body, their toxicity, non-biodegradability, persistence nature and bioaccumulation were previously well documented (Rahman and Singh, 2019; Garg *et al.*, 2007). During the leather manufacturing process, several chemicals like Cr $(SO^4)_3$, NaCl, Ca $(OH)_2$, H_2SO_4, etc. are extensively used. Therefore, the resultant effluent is enriched with chromium and sodium salts (NaCl and SO_4) (Mandal *et al.*, 2011). The investigation became necessary to check the flow of Chromium and other heavy metals in the

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region as tanneries grew in number (Kanagaraj and Elango, 2019). In these areas, although tannery industries that did not have their own effluent treatment plants or not connected to a shared effluent treatment facility were already closed, still chromium contamination exist in the oxidation states of +2, +3, and +6, among these forms, owing to limited water solubility, the trivalent forms remain as a more stable form in the soil system (Sharma *et al*., 2021). The pH of the solution, the total Cr content, the presence of oxidizing and reducing chemicals, the redox potential, and the kinetics of the redox reactions all influence this distribution (Dhal *et al*., 2013).

A considerable number of small-scale tanneries across the country lack access to a common treatment plant, forcing them to dump their waste in open fields or landfills (Manoj Kumar *et al*., 2018). Sludge is also commonly used in agriculture activities as readily available and affordable manure. These improper disposal methods contaminate the soil and water, making it easy for Cr to infiltrate the food chain. Plant chromium absorption accounted for less than 1% of chromium eliminated from the soil (Banks *et al*., 2006). Cr (VI) is easily converted to Cr (III) once ingested by plants (Aldrich *et al*., 2003). The addition of organic materials had the most significant impact on chromium mobility overall (Sathya *et al*., 2021). Hexavalent chromium [Cr (VI)] issued in a variety of sectors, including stainless steel, electroplating, dyes, and leather tanneries, as well as in wood preservation procedures (Jobby *et al*., 2018). The disposal of chromium-contaminated sludge - produced as a by-product of wastewater treatment (Wang *et al*., 2021) is one of the primary rising environmental challenges in the tanning industry (Chatterjee *et al*., 2011). It penetrates many environmental systems (air, water, soil, etc.) through either natural or anthropogenic processes such as mining, smelting operations, metal processing, industrial production, residential and agricultural metal use (Sandeep *et al*., 2019). Heavy metals and some organic contaminants can accumulate in the body, causing cancer, reproductive problems, and other serious health problems. As a result, understanding the level of contamination and the soil condition becomes critical to protect the valuable agricultural soil resources for future generations (Sarwar *et a*l., 2017). The Cr concentration in soil is markedly higher than the average value reported in different parts of the Vellore district (Rangasamy *et al*., 2015; Sinduja *et al*., 2022). The accumulation of Cr in the soil is of concern because of its possible phytotoxicity or increased movement of metals into the food chain and the potential for surface and groundwater contamination. Chromium contamination in soil and water has drastically reduced the crop yields (25 to 40%) over the years and decreased significantly (Mahimairaja *et al*., 2011). Because of the possibility of phytotoxicity or increased metal transport into the food chain and the potential for surface and groundwater contamination, the accumulation of Cr in the soil is a source of concern (Li *et al*., 2019). Chromium contamination in soil and water has resulted in a considerable reduction in crop yields (25 to 40%) and a significant loss in crop quality (Mahimairaja *et al*., 2011). The primary goal of this project is to examine several physicochemical characteristics of the soil in Walajapet, Tamil Nadu, India, which is the hub of tannery industries (Nazir *et al*., 2015).

MATERIAL AND METHODS

Study Area

The Walajapet Taluk is located at Northern latitude and 93 km west of Chennai. It is geographically 25 km away in the North East of Vellore. The site chosen for the present study was the SIPCOT industrial complex area near Walajapet. The tannery is the basic unit of the leather industry, which requires a large amount of water and chemicals and major parts are discharged as effluent. There are more than 200 Tanneries and finished leather goods manufacture industries located in near the Walajapet Taluk of Vellore District, Tamil Nadu, India.

Soil sample collection

Soil samples were collected from chromium-contaminated sites. First, surface soil samples were collected from 0 – 15 cm depth. Then the soil samples were then dried in the shade for 2-3 days ground and sieved (2 mm sieve) and packed in polyethylene cover, and used for further analysis. Finally, the analysis of various parameters was carried out as per the standard methods characteristics of the soil samples (Table. 2).

Total Chromium

By digesting the soil sample with aqua-regia, the total Cr concentration in the soil sample was determined. In a 100 mL conical flask, one gram of soil sample was weighed, and 15 mL of aqua-regia (HCl: HNO_3 at 3:1 ratio) was added and digested on a hot plate till white fumes evolved. The digestate was diluted to 100 mL of water and filtered through Whatman No. 42 filter paper. Using Atomic Absorption Spectroscopy (Varian Spectra, AA220) and an air acetylene flame at 357.9 nm, the total Cr contained in the solution was determined (USEPA, 1979).

Hexavalent Chromium

The hexavalent Cr concentration in soil was determined using the Diphenyl carbazide method (USEPA, 1979). Diphenyl carbazide reagent was prepared by dissolving 0.25 g of 1, 5 - diphenyl carbazide, and 4 g of phthalic anhydride in 100 mL of 95 percent ethyl alcohol.

One gram of the soil sample was taken in a 50 mL polypropylene centrifuge tube, and 25 mL of distilled water was added. The contents were shaken using an end-over-end shaker for 2 hours, centrifuged at 8000 rpm for 10 minutes, and filtered using Whatman No.1 filter paper. The 10 mL of extract was taken in a 50 mL volumetric flask. To this, 10 mL of 1N H_2SO_4 and 4 mL of 1, 5-diphenyl carbazide reagent were added, and the volume was made up to 50 mL with distilled water. The content was allowed to stand for 20 minutes for purple – violet color development. The absorbance of the color was measured at 540 nm using a Spectrophotometer

Trivalent Chromium

The Cr (III) can be determined using the below equation (Zhao *et al.*, 2019)

$$Cr\ (III) = total\ Cr - Cr\ (VI)$$

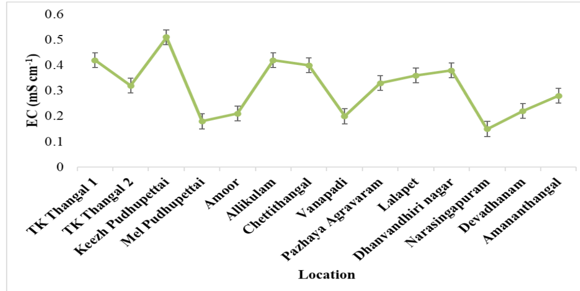

Figure 1. EC variations of Chromium content of soils collected from Walajapet Taluk of Vellore district

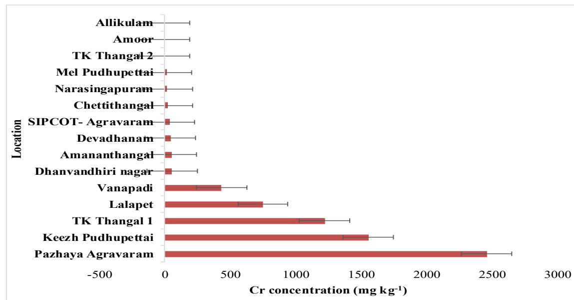

Figure 2. Chromium content of soils collected from Walajapet Taluk of Vellore district

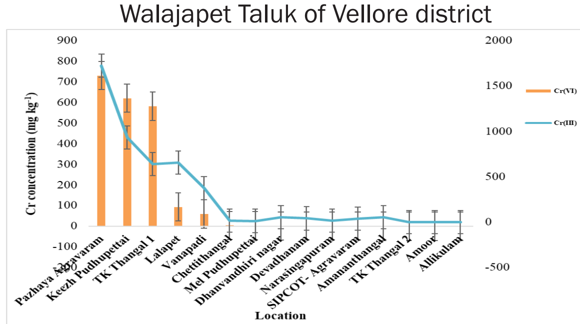

Figure 3. Chromium speciation contamination in the villages of Walajapet Taluk of Vellore district

Table 1. Geographic sample locations and physical parameter analysis

Sample No	Location	Latitude	Longitude	pH	EC (mS cm^{-1})
1	TK Thangal 1	12°54'59"N	79°22'52"E	8.14	0.42
2	TK Thangal 2	12°56'10" N	79°21'15" E	8.20	0.32
3	Keezh Pudhupettai	12°56'25" N	79°22'44" E	8.14	0.51
4	Mel Pudhupettai	12°56'25" N	79°22'44" E	8.16	0.18
5	Amoor	12°58'36" N	79°21'54" E	8.72	0.21
6	Allikulam	12°57'33" N	79°22'65" E	8.10	0.42
7	Chettithangal	12°57'18" N	79°20'62" E	8.55	0.40
8	Vanapadi	12°57'44" N	79°19'46" E	8.54	0.20
9	Pazhaya Agravaram	12°58'17" N	79°18'45" E	8.46	0.33
10	Lalapet	12°32'11" N	78°42'47" E	7.18	0.36
11	Dhanvandhiri nagar	12°57'72" N	79°22'13" E	6.66	0.38
12	Narasingapuram	12°36'32" N	78°44'29" E	8.25	0.15
13	Devadhanam	12°55'47" N	79°21'64" E	7.45	0.22
14	Amananthangal	12°54'75" N	79°22'29" E	7.62	0.28
15	SIPCOT- Agravaram	12°58'62" N	79°18'30" E	7.50	0.38

Table 2. Threshold levels of Cr permissible in soils (mg kg^{-1}) of some developed countries

Countries	Metals	Cr (mg kg^{-1}) permissible limit	Reference
UK	Cr (III)	600	UK Department of Environment (1987)
	Cr (VI)	10	
Canada	Cr (III)	250	CCME (1990)
	Cr (VI)	8	
USEPA	Cr (III)	1500	USEPA (1993)
	Cr (VI)	-	
Australia	Cr (III)	50	ANZECC / NHMRC (1992)
	Cr (VI)	-	

RESULTS AND DISCUSSION

Soil chemical parameters

Around 64 surface soil samples from the Vellore district were analyzed for physical parameters shown in Table. 1. According to the results obtained in Walajapet Taluk, soil pH ranged from 6.66 to 8.55, with an average value of 7.9, signifying alkalinity in the study region. The level of pH concentrations does not exceed the permissible limits (6.5-8.5) recommended by WHO and IS10500. The higher value recorded in the chettithangal village was due to the tannery effluent and geochemical processes, the soil samples near the tannery and rock weathered region had a higher pH and EC, and the presence of calcareous nature in the study area is a major reason for the elevated level of pH. A pH of more than 8.0 was found in around 70% of the samples, indicating soil sodicity. EC is an indicator of salt accumulation; EC ranged from 0.15 to 1.38 mS cm^{-1} was found lower and tabulated in Table. 1. Fifty percent of soil samples examined were found to have EC values of more than 1 mS cm^{-1}, and it was represented in Figure 1. The substantial fluctuations in EC and higher EC in the study area are mainly attributed to anthropogenic activities and the nearby tannery region influence of tannery effluents. The concentration of sodium increases in the soil, which is due to the geochemical processes of silicate weathering and anthropogenic activities. Increased EC can be linked to higher salinity and mineral content in soil samples generated by geochemical processes such as ion exchange, evaporation, silicate weathering, and solubilization in the study region (Ramesh and Elango, 2012). EC generally rises due to the combined impacts of evaporation, ion exchange, and groundwater flow path (Tóth, 1999). The pH and EC values of soil samples near the tannery and rock weathering zone were higher due to tannery wastewater.

Total Chromium

Chromium contamination was shown in Figure 2; the chromium was higher in the soil of Pazhaya Agravaram (2459 mg kg^{-1}) of Walajapet, followed by Keezh Pudhupettai (1556 mg kg^{-1}) village. Compared to all the villages, Walajapet showed a higher chromium concentration above the permissible limit described in the Table. 2. The data clearly showed that the soil close to tannery industries is severely contaminated with Cr., was mainly due to the operation of the tannery industries, as they contaminate the surface soil with heavy metals like chromium, manganese, copper, lead, cadmium, and nickel (Uddin and Ahmed, 2018). Because tannery effluent released on land, particularly at high concentrations (100 percent), enhanced chromium content in the soil, higher levels of Cr were discovered near the research region (Rangasamy *et al*., 2015). Industrial wastes such as Cr pigment, raw tannery wastes, and industrialized leather wastes are responsible for the high concentration of Cr in the tannery surrounding area topsoil samples. Cr pollution in the topsoil set down in the soil as well as in the groundwater due to the tanning industries area of Ranipet and Walajaphet town, Vellore city. The tanneries discharge untreated tannery effluents, which get mixed with the soil, water of the Palar river, and underground in this area (Kistan and Kanchana, 2020). Cr content showed no definite pattern in soil but tended to accumulate in disproportionate amounts in the soil. The difference could be attributed to differences in soil texture and hydrological aspects of sampling sites, which could have altered Cr mobility, resulting in varying amounts of Cr in surface and subsurface soil. The different chemical makeup of tannery effluent and sludge could have also influenced Cr accumulation. The findings support those of (Sinduja *et al.*, 2022), who recently found severe soil contamination in Vellore due to Cr from tannery wastes.

Hexavalent and Trivalent Chromium

Chromium (VI) and chromium (III) are under total chromium, whereas chromium (VI) is highly toxic and chromium (III) has less toxicity. Hexavalent chromium is one of the oxidation forms of chromium, which leads to lung cancer, chromate ulcer, perforation of the nasal system, and kidney damage in humans and other organisms (Sunitha and Rajkishore, 2013). In this study, hexavalent chromium levels exceed the permissible limits as per WHO. Therefore, disposal of hexavalent chromium contaminated water to the stream cause a serious issue of groundwater contamination. In the soil, the Cr (VI) level ranged from 0.20 to 731.5 mg kg^{-1}, as shown in Figure 3. Pazhaya Agravaram (731.5 mg kg^{-1}) had the most bioavailable Cr, followed by Keezh Pudhupettai (621.1 mg kg^{-1}). Cr (III) concentrations in the studied region ranged from below detectable level to 1727.5 mg kg^{-1}. A higher trivalent concentration was found in the Pazhaya Agravaram village of Walajapet Taluk. Chromium can appear in both trivalent Cr (III) and hexavalent Cr (VI). Hexavalent chromium is one of the oxidation forms of chromium, and it is far more hazardous than trivalent chromium. In humans, exposure to hexavalent chromium causes lung and stomach cancer. The hexavalent chromium was the higher limit in the soil sample in the present study. On all other sampling sites, the concentration was higher at 731.5 mg Kg^{-1}, which is much above the limit prescribed by WHO standards (Table. 2).

Furthermore, soil and sediments and aquitard sediments will substantially impact soil chemistry since they indicate the soil's passage pattern (Shahid *et al*., 2017). Under acidic pH, Cr (III) gets reduced to Cr (VI), which gets leached with acidic rainwater (Nouri and Noroozifar, 2018). The decline in soil Cr could be attributable to the oxidation of Cr (III) to Cr (VI), which dissolved in water and leached down into the soil profile, eventually reaching groundwater over ten years (Thangam *et al*., 2018). Humans require chromium-III as a dietary supplement, and it is frequently added to vitamins. Chromium (III) has low toxicity and would only be a problem in drinking water if contamination levels were high. Chromium (VI) is more poisonous and might harm your health. Over an extended period, those who drink water with total chromium levels above the permissible limit contamination threshold may develop allergic dermatitis.

CONCLUSION

The study results show that the soil in the Walajapet area is significantly contaminated by Cr, which may give rise to various health problems or diseases. The detected levels of total chromium contamination in many of the samples were revealed that the majority of soil samples collected from 15 places in Walajapet -Taluk exhibited high levels of Cr (>200 mg kg^{-1}), above the permissible (threshold) limit set by WHO in various nations, particularly in India. High Cr values confirm that the contamination is originating from the tannery waste. Total chromium showed a strong anthropogenic influence. Soil EC indicates agricultural return flow and chemical seepage. In the northern part of the research area, there was a significant concentration of Cr (III) and Cr (VI). The significant metal level in soil samples near tanneries in the Walajapet area demonstrates the impact of tannery effluents. As a result, it is essential to develop strategies to improve the effluent treatment plant functional efficiency to prevent the improper discharge of untreated wastewater into open drains and to implement appropriate remediation technologies for chromium removal from agricultural lands and affected areas. Such innovations would assist and keep agriculture viable, and they would also help mitigate adverse effects on the environment. The area requires remediation as per environmental quality criteria and regular monitoring of toxic metals enrichment.

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Ethics statement

No specific permits were required for the described field studies because no human or animal subjects were involved in this research.

Originality and plagiarism

Authors should ensure that they have written and submit only entirely original works.

Consent for publication

All the authors agreed to publish the content.

Competing interests

There were no conflict of interest in the publication of this content

Data availability

All the data of this manuscript are included in the MS. No separate external data source is required. If anything is required from the MS, certainly, this will be extended by communicating with the corresponding author through corresponding official mail.

Author contributions

MS: Formal analysis, Investigation, Data Curation, Writing - Original Draft, Visualization, Software analysis, Conceptualization, editing; VS: Conceptualization, Methodology, Validation, Review & Editing, Project administration, Funding acquisition, Supervision; MM: Supervision, Review & editing, Investigation, Editing; PD: Review & editing, Validation; TC, US: Review & editing; PK: Resources, Lab accommodation, Review & editing.

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