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HEAVY METAL CONTAMINATION IN WATER, SOIL AND VEGETABLES OF TANNERIES POLLUTED AREA OF VANIYAMBADI, TAMIL NADU, INDIA

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ABSTRACT

Tanneries discharge waste water containing chemicals including chromium salt. This effluent contaminates the soil and water bodies nearby. Therefore, levels of Cr along with Cd, Cu, Ni, Pb and Zn metal ions were analyzed in groundwater, soil, vegetables and fruits harvested around the tannery areas of Vaniyambadi, Tamil Nadu. Analysis results showed that the mean levels of Cr in the soil and ground water samples exceeded the acceptable limit. Heavy metals' accumulation in vegetables was found to be below the permissible levels proposed by the Indian standard with the exception of Cr which exhibited an elevated level in some of the vegetable samples. Generally, the Cr levels in water, soil, vegetables and fruits that are grown in proximity to the tanneries greatly exceeded the background levels indicating an extreme degree of chromium contamination. Comparison of the heavy metal levels with those of the established safe limit levels provides further evidence for proper treatment and regular monitoring of toxic metals before their discharge into the environment.

Keywords: Tannery, Environment, Groundwater, Vegetables, Heavy Metals, Pollution

INTRODUCTION

The leather industry makes a significant contribution to India's foreign exchange earnings and employs about three million people. In the tanning process, animal skins/hides are transformed into leather in successive treatments using large quantities of water and many chemicals like NaCl, $Ca(OH)_2$, H_2SO_4 , $Cr(SO_4)_3$ etc. A considerable volume of chemical loaded waste water is generated and discharged as effluent. The major chemical affecting the ecosystem is the salts of chromium. This is characterised in the effluent by its strong colour (reddish dull-brown), high levels of BOD, pH and total dissolved solids (TDS).

Tannery effluents, being voluminous and highly contaminated, when discharged untreated, damage the normal life of the receiving water bodies; and if they percolate into the ground for a prolonged period, will pollute the groundwater permanently and make it unfit for drinking, irrigation and domestic consumption. Groundwater is the main source of water supply for domestic use and drinking purpose in India's rural areas. A single tannery can cause the pollution of groundwater within a radius of 7–8 km (Bhaskaran, 1997; CLRI, 1990; Ansari *et al.*, 1999). Chromium in effluent at its generation point is mainly a less toxic trivalent form Cr (III), but when this enters the soil, Cr (III) is gradually oxidised to the more toxic hexavalent form through varying environmental conditions (Anderson, 1999; Govil *et al.*, 2004; Gowd *et al.*, 2005).

Since tannery effluent contains toxic chemicals which can have serious effects on biota and the environment, proper pre-treatment is essential. Yet small industries occasionally release their effluent directly into the environment (Kannan and Uperti, 2007). For various reasons, in many areas of Tamil Nadu, where tanneries are located, groundwater is not suitable for domestic use and therefore villagers (women and children) are forced to travel 4–5 km for water. Much of the groundwater is unsuitable for

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irrigation and hundreds of wells in the region are no longer in use. Since the heavy metal pollution of agricultural soil contaminates the food chain, this is the major pathway of heavy metal exposure to human beings (Khan *et al.*, 2008). Some trace elements are essential in plant nutrition, but plants growing adjacent to industrial areas display an increased concentration of heavy metals, thus serving as biomonitors (Mingorance *et al.*, 2007).

Vegetables cultivated in soil, polluted with toxic metals through industrial activities, take up heavy metals and accumulate them in their edible and non-edible parts in quantities high enough to cause clinical problems both to animals and human beings. Once accumulated in the human body, there is no effective mechanism for their total elimination (Arora *et al.*, 2008; Alam *et al.*, 2003). Trace elements are very harmful because of their non-biodegradable nature, long biological half-lives and their potential to accumulate in different body parts.

Many studies have reported on the deposition of heavy metals in soil, crops and vegetables grown in the vicinity of industrial areas (Grytsyuk *et al.*, 2006; Mingorance *et al.*, 2007). The Agency for Toxic Substances and Disease Registry (ATSDR) succinctly characterises the adverse health effects information for Chromium.

Hexavalent chromium is a toxic industrial pollutant and classified carcinogen, being mutagenic and teratogenic. Absorbed chromium can be transferred to foetuses through the placenta and to infants via breast milk (Niyas *et al.*, 2014).

To the best of our knowledge, levels of different heavy metals in the area of Vaniyambadi, particularly the chromium in soil, vegetables and water samples from the tannery there, has not previously been reported. Accordingly, and to extend our previous work (Sujatha *et al.*, 2013; Sandana Mala *et al.*, 2015) we investigated the level of Cr, Cd Cu, Ni, Pb and Zn in groundwater, soil, vegetables and fruits from agricultural land near a tannery located at Vaniyambadi, Vellore District, Tamil Nadu, India. The aim was to highlight the contamination status of heavy metals in the edible portion of vegetables grown in soil polluted by the tanning industry in Vaniyambadi municipal area and to correlate the metal concentrations of that polluted soil with that of the crops.

MATERIALS AND METHODS

Study Site Description

The selected study area, Vaniyambadi, the Leather City of South India, located about 200 KM from Chennai, part of Vellore district of Tamil Nadu, India is chronically polluted. It is one of the hubs of leather exports in Tamil Nadu (Figure. 1). There are 124 tanning industrial units located in and around this town; there are also brick and ceramic industries (CLRI, 2013).

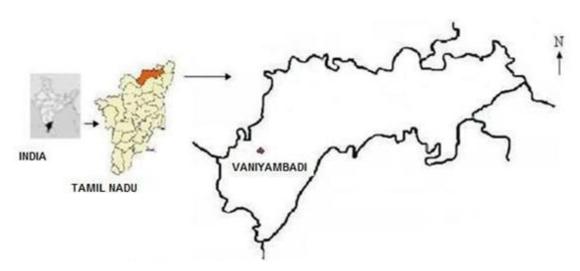


Figure 1: Vellore District with the Study Area

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Sampling, Storage and Analysis

Soil, water and vegetable samples for the study were collected from areas adjacent to selected tanneries of Vaniyambadi, the study area. The experiment consisted of analyses of heavy metals present in soil, water and some of the farm products of the locality.

Soil

Individual soil samples of 10 x 10 x 15 (L x W x D) cm monolith were collected in triplicate, air dried, crushed, and sieved through a < 100 μ sieve, oven dried at 80°C for 48 hours and stored in polythene bags for sample analysis.

Water

Water samples from five different wells were collected to assess the extent of heavy metal contamination. These samples were collected in 1L capacity, double cap polythene bottles and transported for laboratory analysis. Prior to collection the sample bottles were thoroughly washed with acid and then with distilled water.

Vegetables Sampling

Vegetable samples from the study area were collected in zip lock bags after hand harvesting and transported to the laboratory under cold conditions. Then, the samples were washed with double distilled water to remove airborne pollutants, and oven dried at 80°C until the moisture was removed. Dried samples were powdered using a pestle and mortar and sieved.

Processing of Sample

Soil and Vegetable Samples

A 0.5g moisture free soil or vegetable sample was digested on a hot plate with 15mL of digestion mixture containing HNO_3 : $HClO_4$ in the ratio of 6:1 until white fumes appeared in the flask (NIOSH, 1987).

Water Sample

Acidification of 200mL of collected water sample occurred by the addition of 10 mL of concentrated HNO₃. The sample was digested on a hot plate until the volume was reduced to 2-3 mL. The solutions were then filtered separately through Whatman No.1 filter paper and the filtrate was made up to 10 mL with 0.1 N HNO₃.

Quantification of Heavy Metals in Soil, Water and Vegetables

The concentrations of metals in the processed samples were determined by using atomic absorption spectrophotometer (Perkin Elmer, USA, model Analyst 300). Blanks containing only distilled water were prepared and processed in the same manner along with all types of samples. All the reagents used were of Merck, analytical grade.

RESULTS AND DISCUSSION

Heavy Metal Content in Soil

Continuous discharge of chromium containing effluent from tanning industries led to accumulation of significantly higher concentrations of heavy metals in soil at the contaminated site. The soil metal content other than chromium was found to be either below or within the usual typical background values (Awasthi, 2000) (Table 1). The average concentrations of Cr, Cd, Cu, Ni, Pb and Zn analysed were 17.85, 0.41, 20.68, 1.93, 2.97 and 93.8 mg/kg at 35 metres, 11.71, 0.38, 9.42, 0.141, 3.79 and 54.61 mg/Kg at 500 metres and 2.545, 0.08, 0.072, 0.943, 0.024 and 0.14 mg/Kg at 1000 metres, respectively from the discharge point. The concentrations of heavy metals below the safe limits around the activity area may be caused by the leaching of certain metals into the deeper layers of soil or the continuous absorption of these metals by the agro plants grown in this area.

The concentration of metals is decreased when effluent travels a long distance in the soil, from the point of its origin. Moreover, an increased chromium level may be attributed to the poor solubility of basic chromium sulphate (BCS) or its restricted absorption by plants caused by transportation barriers. BCS is used in tanning activity at acidic pH, for enhanced solubilisation and penetration in the leather forming substrate, the pelt. When the effluent from tanning is combined with other effluents with elevated pH, the solubility and absorbability of Cr is affected.

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Table 1 presents the mean chromium concentrations of three replicates in the soil along with other heavy metals of each sampling station. It was observed from the results that the Cr and other metal concentration in the soil sample collected 35 metres away from the tannery is higher than those found from soil samples collected from 500 and 1000 metres away. When plants grow on metal contaminated soil, they accumulate the metals in their different parts, sometimes beyond their tolerance level resulting in reduced yields because metabolic processes have been inhibited. Even the growth of the plant itself is likely to be adversely affected (Sanders *et al.*, 1987). Loading of heavy metals often leads to degradation of soil health and contamination of the food chain, mainly through the vegetables grown on such soil (Jackson and Alloway, 1992; Rattan *et al.*, 2002). The observed increase in Cr content of the soil of Vaniyambadi is attributed mainly to the chrome tanning activity in the area and discharge of Cr-containing effluents.

Heavy Metal Content in Groundwater

The groundwater samples of the soil contaminated regions were tested, presuming the penetration by pollutants from surface to subsoil layers and into the groundwater. The water samples collected at different locations of Vaniyambadi showed wide variations in their heavy metal content. Out of six elements examined in the groundwater collected from locations near the tanneries at Perumalpet and Valayampet area of Vaniyambadi, Cr recorded the highest level and exceeded the permissible limit. The other metals exhibited a lower concentration than the permissible levels of drinking water specifications (IS 10500:2012). The normal range of chromium in irrigation water is 0.1mg/L (Pescod, 1992; USEPA, 2003) while the normal range of Cr in drinking water is 0.05 mg/L.

Although, Cr (III) is considered as an essential trace element for the maintenance of an effective glucose level, lipid and protein metabolism in mammals, Cr (VI) is toxic for biological systems. It is water soluble and is extremely irritating and toxic to human body tissue owing to its oxidising potential and the permeability of biological membranes (Ansari et al., 1999). Chromium levels in the study area ranged from 0.13 to 1.87 mg/L with an average of 0.836 mg/L of water (Table 2). The levels of five other heavy metals, namely Cd, Cu, Pb, Ni and Zn, are within the permissible limits. Particularly, Cd and Ni levels were negligible in the water samples tested. This implies that the distribution of heavy metals and particularly Cr in Vaniyambadi area is mainly caused by tannery activity. Heavy metals in groundwater are also associated with small scale industries such as colouring, electroplating, metal surface treatments, fabric painting, battery and paints releasing Cr, Cu, Ni, Pb and Zn. The lower concentrations of heavy metals other than Cr in the water sample of this study may be caused by dilution in the water medium. But, the discharge of tannery effluents with an excessive Cr concentration into fresh water sources and dumping of tannery wastes leads to the contamination of groundwater and the surface soil. Cultivation of crops in the polluted land and irrigation using polluted groundwater with a high concentration of heavy metals eventually causes the entry of heavy metals into the food chain. It is disturbing to note that the use of contaminated drinking water will pose a health risk to the people dependent on groundwater at these sites. Gowd and Govil (2008) have reported an increased level of heavy metals in groundwater in the tanning industry active at Ranipet which made the groundwater unsuitable for domestic use, forcing villagers to travel far to collect potable water. Much of the groundwater is declared unsuitable for irrigation and hundreds of wells in this region have been reported to be unfit for domestic use (Thangarajan, 1999; Selvakumar and Manoharan, 2002; Mondal et al., 2005). Because of their characteristic toxicity and persistence in biological systems, the heavy metals are aquatic pollutants which subsequently enter the food chain through absorption by plants (Uysal et al., 2008).

Heavy Metal Content in Vegetables

Heavy metal pollutants from industrial sources travel through soil and water to reach the plants. Agricultural activity near the polluted area results in absorption of the toxic metals from soil/water to accumulate them in the plant parts, both edible and inedible. Thus, Table 3 records measurement of accumulated heavy metals in some of the food grains, fruits and vegetables harvested from the Vaniyambadi area. Heavy metal contamination of food items has been reported in India, Zambia, Egypt and China (Marshall *et al.*, 2007; Radwan and Salma, 2006; Wang *et al.*, 2005). The mean concentrations of heavy metals in test vegetables showed many variations.

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Table 1: Metal Concentration in Soil Collected near the Tannery of Vaniyambadi Town

Sampling Location (from Tannery Industrial Area)	Metal Concentration (mg/Kg)							
	Cr	Cd	Cu	Ni	Pb	Zn		
35 meters	17.85±0.56	0.41±0.1	20.68±0.136	1.93±0.061	2.97±0.105	93.8±0.12		
500 meters	11.71±0.09	0.38 ± 0.006	9.42±0.029	0.141 ± 0.001	3.79 ± 0.0024	054.61 ± 0.78		
1000 meters	2.545±0.02	0.08±0.0003	0.072±0.003	0.943±0.0035	0.024±0.004	0.14±0.001		
Safe limit ^a		3-6	135-270	75-150	250-500	300-600		

Data represents Mean±SD (n=3)

Table 2: Metal Concentration in Water Samples Collected near Tanneries of Vaniyambadi Town

Well Water Sample	Metals (mg/L)							
Location	Cr	Cd	Cu	Ni	Pb	Zn		
Perumalpet-1	1.45±0.006	0.004±0.002	0.01±0.014	0.03±0.025	0.004 ± 0.002	1.07±0.002		
Perumalpet-2	0.23±0.0008	0.005 ± 0.001	0.006 ± 0.004	0.02 ± 0.003	0.005 ± 0.001	0.2 ± 0.001		
Perumalpet-						0.12±0.009		
Extension	1.87 ± 0.0002	0.0003 ± 0.0001	0.01 ± 0.006	0.03 ± 0.015	0.0003 ± 0.0001			
Valayampet-1	0.50 ± 0.0003	0.003 ± 0.002	0.02 ± 0.004	0.01 ± 0.001	0.003 ± 0.002	0.066 ± 0.048		
Valayampet-2	0.13±0.0002	0.004 ± 0.001	0.006 ± 0.002	0.012 ± 0.001	0.004 ± 0.001	1.16±0.010		
Water quality criteria for irrigation ^a	0.1	0.01	0.2	0.2	0.5	2.00		
Safe Limit IS-10500:2004 ^b	0.05	0.003	0.05	0.02	0.01	5.0		

Data represents Mean±SD (n=3)

^a Source: Awasthi (2000)

^a Source: Pescod (1992)

b- Drinking water specifications (Indian standard)

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Table 3: Metal Concentration in Fruits/Vegetables Collected from Locations near Tannery Area of Vaniyambadi

Type of Fruits/	Metals (mg/Kg)			•		
Vegetables	Cr	Cd	Cu	Ni	Pb	Zn
Goosebery	10.25±0.095	0.84±0.081	4.61±0.100	0.15±0.095	1.63±0.095	23.42±0.084
Brinjal	59.33±2.081	1.34 ± 0.105	10.43±0.090	1.25 ± 0.068	17.01±0.115	55.25±0.068
Pomegranate	21.46±0.101	0.37 ± 0.009	5.52 ± 0.090	0.86 ± 0.105	1.44 ± 0.080	16.26±0.105
Custard Apple	9.06 ± 0.062	0.43 ± 1005	4.41 ± 0.095	1.04 ± 0.105	1.13±0.060	46.28±0.100
Sugar Cane	3.33±0.153	0.46 ± 0.208	12.46±0.110	1.47 ± 0.102	5.04 ± 0.120	37.46±0.251
Groundnut	2.42±0.080	0.15 ± 0.095	10.22±0.209	1.07 ± 0.015	0.67 ± 0.095	6.62±0.095
Maize	33.42±0.110	0.08 ± 0.007	1.92 ± 0.055	0.42 ± 0.107	0.21 ± 0.108	6.11±0.111
Corn	3.42±0.105	0.07 ± 0.015	2.13±0.120	0.42 ± 0.106	0.24 ± 0.072	6.11±0.102
Coconut	84.97±0.104	0.28 ± 0.096	9.21±0.107	1.45±0.112	0.55 ± 0.115	21.94±0.080
Rice	2.76±0.115	0.15 ± 0.130	2.05 ± 0.064	0.47 ± 0.100	0.45 ± 0.085	12.65±0.101
Toordal	3.12±0.105	0.21 ± 0.095	3.85 ± 0.081	1.38 ± 0.095	0.41 ± 0.098	17.88±0.095
Ragi	57.63±0.37	0.18 ± 0.091	8.23±0.157	ND	1.74 ± 0.015	10.06±0.153
Papaya	41.56±0.351	1.33 ± 0.208	3.41 ± 0.152	1.44 ± 0.045	ND	67.96±0.080
Drumstick leaves &						
Flower	11.33±0.015	0.55 ± 0.095	2.47 ± 0.110	0.45 ± 0.030	ND	37.53±0.152
Sweet Tamarind	2.21±0.049	0.21 ± 0.074	3.41 ± 0.097	0.06 ± 0.015	ND	14.35 ± 0.095
Safe limit ^a	20.00	1.5	30.00	1.5	2.5	50.00
Safe limit ^b	5.0	0.3	40	20	5.0	60
Safe limit ^c	0.5	0.2	20	10	9.0	100

Data represents Mean \pm SD (n=3).

^a Source: Awasthi (2000) ^b FAO/WHO standard (Codex Alimentarious Commission 1984)

^c State Environmental Protection Administration, (SEPA,China, 2005)

ND: Not detected, Levels were below the detection limit

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The metal accumulation in the farm produce was found to be in the order of Zn>Cr>Cu>Pb>Ni>Cd. The level of accumulation of metals, other than Cr is in the range of 6.11- 55.42 for Zn; 1.92-10.22 for Cu; 0.21-17.0 for Pb; 0.15-1.47 for Ni and for Cd 0.15-1.34 mg/Kg. The concentration of Cr in vegetables/fruits is ranging widely from 2.21 to 84.97 mg/Kg. The highest level of Cr accumulation was found in coconut (84.97 \pm 0.104mg/Kg) followed by brinjal (59.33 \pm 2.08 mg/Kg), ragi (57.63 \pm 0.37 mg/Kg) and maize (33.42 \pm 0.11mg/Kg). The highest level of accumulation of Zn was recorded by papaya (67.96 \pm 0.08 mg/Kg), Cu by groundnut (10.22 \pm 0.020 mg/Kg), Pb by brinjal (17.01 \pm 0.11 mg/Kg) and Cd by brinjal (1.34 \pm 0.105 mg/Kg). Differences in the levels of Cr and other metals in vegetables imply that different types of vegetables/plants have different mechanisms to absorb and accumulate the metal ions. Various investigators have reported the differential absorption capacity of vegetables for different metals (Zurera *et al.*, 1989) and their further translocation within the plants (Voutsa *et al.*, 1996) and the translocation factor of each metal (Cui *et al.*, 2004; Zheng *et al.*, 2007).

The observed values of Cr in the present study exceed the average normal concentration and are beyond human safe tolerance level. This may create serious health problems in the long run. In our previous study, we found that the maximum accumulation of Cr was in papaya ($52.36 \pm 0.105 \text{mg/Kg}$) followed by Citrus ($34.44 \pm 0.032 \text{ mg/Kg}$) collected from Ranipet industrial area of Vellore District, Tamil Nadu.

In this study, increased levels of chromium were observed in soil, water and vegetable samples collected from the site/farm in close proximity to tanning industries. These increased levels are attributed to the tanning industries' discharge of Cr containing waste water into the surrounding area. In our earlier work, we made a similar observation on the heavy metal accumulation profile in the target sources of State Industries Promotion Corporation of Tamil Nadu (SIPCOT) industrial area of Ranipet, India (Sujatha *et al.*, 2013). The transfer of heavy metals from the tannery chemical sludge to soil and the subsequent uptake from soil and accumulation of the heavy metals in the edible parts of vegetative tissue allow them to enter the human body by normal processes of consumption of foodstuffs (Dowdy *et al.*, 1984).

Recently, Sharma *et al.*, (2009) carried out a health survey and clinical examination on community inhabitants from the big leather working centre of Kanpur, India where groundwater is contaminated with hexavalent chromium owing to tannery activities and chrome sulphate manufacturing industries. In Kanpur and surrounding areas, gastrointestinal (GI), ocular complaints and overall lung function abnormalities remained significant among exposed males. In females, urinary complaints with GI and dermal disorders were significantly higher. Besides, some significant difference in haematology parameters such as Red blood corpuscles (RBC), Platelets (PLT) and Mean cell volume (MCV) has also been reported among the exposed subjects. In the tannery area of Ranipet, South India, occupational diseases such as asthma, chromium ulcers and skin diseases have been observed (Mahimairaja *et al.*, 2000; Gnanasekaran *et al.*, 2010).

Conclusion

Based on the findings of this study we conclude that the Cr accumulation is highest in the soil followed by groundwater and then vegetables. Thus, accumulated heavy metals would cause a health risk to all biota. High levels of Cr in the ecosystem confirm that the contamination was mainly caused by the release of uncontrolled effluents or dumping of industrial wastes on to the ground which pollutes the soil and water bodies in the study area. Based on the Writ Petition (C) No. 914 of 1991 by Vellore Citizens Welfare Forum, the Supreme Court of India observed that the leather industry was of vital importance to the country as it generated foreign exchange and provided employment avenues. But, the Court added that tanneries had no right to destroy the ecology, degrade the environment and cause a health hazard. In obedience of the direction of the Supreme Court, the Loss of Ecology (Prevention and Payment of Compensation) Authority has been constituted by the Government of India, vide its Notification in S.O.671 (E), dated 30.9.1996 with a retired Judge of this Court, as the Chairperson and the said Authority identified 29,193 individuals/families as affected. Accordingly, 546 tanneries in the District of Vellore were ordered to pay a compensation amounting to INR 26.82 crores [INR 26,82,02,328/-] (US\$ 3.8 million) to the affected as pollution damages (Vellore Citizens Welfare Forum v. Union of India and Ors. MANU/SC/0686/1996).

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There is an urgent need to monitor regularly the levels of toxic metals in industrial effluents before releasing them; whether from an individual industrial plant or from a Common Effluent Treatment Plant (CETP). Such monitoring should ensure that the load of these toxic metals is minimised so as to comply with the discharge norms. Additionally, necessary treatment processes should be up to the standard of those in use in developed countries. There should also be periodical monitoring of the bio-accumulation of toxic materials in the farm products, and there should be epidemiology studies to establish facts about the relation of industry, pollutants, consumption of foodstuffs, and human health in an area such as the study site. Release of Cr (III) containing effluent from the tanning industry and treatment of this species of chromium at alkaline pH is likely to convert the less toxic Cr (III) into more toxic and highly reactive Cr (VI).

This Cr (VI), being highly soluble in water compared to its trivalent form, is easily absorbed by plants from contaminated soil and actively transported to the shoot system to be accumulated in fruits and vegetables, leaves and flowers.

This toxic Cr (VI) species is believed to be chemically reduced to a lower oxidation state; but the increased accumulation of this heavy metal has been reported to affect the normal metabolism, growth and productivity of the plants.

It is probable that continuous consumption of such toxic metal species accumulated vegetable/fruits etc. would cause adverse effects in human beings as well. Cr (VI) is considered to be a carcinogen, as this species is a potential reactive oxygen species (ROS) generator. This deleterious species is capable of modifying several biomolecules including protein, lipid and DNA.

Modification of DNA, sometimes leads to over expression of certain biological markers which in turn might trigger the carcinogenesis.

Therefore, in line with the legal decision of India's High Court, legislators, administrators and industrial managers etc should consider applying preventive measures to minimise the toxic metal pollution in nature and to protect the people of India from deadly diseases.

Disclosure Statement

No potential conflict of interest was reported by authors.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the provision of laboratory facilities for the metal analysis by the kind co-operation of Dr. R.C. Murthy and Dr. R.K. Kanojia, Analytical Chemistry Division, CSIR-Indian Institute of Toxicology Research, Lucknow, UP, India.

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