Bull Environ Contam Toxicol (2017) 98:706–711
DOI 10.1007/s00128-016-1983-4

Geo-Accumulation Indices of Heavy Metals in Soil and Groundwater of Kanpur, India Under Long Term Irrigation of Tannery Effluent

M. L. Dotaniya[1] · V. D. Meena[1] · S. Rajendiran[1] · M. Vassanda Coumar[1] · J. K. Saha[1] · S. Kundu[1] · A. K. Patra[1]

Received: 28 September 2016 / Accepted: 18 November 2016 / Published online: 22 November 2016

Abstract Soil and groundwater from long-term (>50 years) tannery effluent irrigated areas of Kanpur were analyzed and significant buildup of heavy metals such as Cr, Ni, Cd, Pb, Zn, and As in the range of 252–972, 23–30, 2.3–14.1, 23.7–58.8, 138–338 and 6.8–11 mg kg^{-1}, respectively in soil was found. Few groundwater samples in the effluent irrigated areas also exhibited high Cr concentration above the permissible limit of United States Environmental Protection Agency. The tannery effluents contained 1.53–57.3 ppm Cr, 0–0.12 ppm Ni, 0–0.02 ppm Cd, 0–0.07 ppm Pb, 0–0.48 ppm Zn and 0–0.03 ppm As. The Geo-accumulation index (I_{geo}) revealed that soil samples were unpolluted to moderately polluted with Cu, Ni, Zn, Pb and As; moderately polluted in case of Cd; and heavily to extremely polluted by Cr.

Keywords Chromium · Geo-accumulation index · Groundwater · Heavy metals · Tannery effluent

Use of waste water for irrigation of agricultural lands in urban and rural areas across the globe is a common practice (Feigin et al. 1991), due to scarcity and unavailability of quality irrigation water. Wastewater use in urban and rural agriculture is increasing where such use derives significant economic activity and supports the livelihood of resource poor farmers. But the major concern is many small scale industries release their effluent straight to surface water bodies without any pre-treatment (Minhas and Samra 2004) because pre-treatment of effluent is very expensive. In the countries where treatment and safe effluent disposal facilities are limited or non-existent, sewage is used to irrigate fodders, ornamental and food crops including vegetables (Ensink et al. 2007). For instances, in India, total waste water generation is 38254.82 million L per day (from 498 class 1 cities and 410 class 2 towns) and available treatment capacity is only 11787.38 million L, rest of the waste water are gets its path into the surface water bodies and soils which causes the contamination (Minhas and Samra 2004; CPCB 2009). Irrigating the agricultural fields with the tannery effluents was found to be beneficial to some extent due to relatively higher load of plant nutrients (Dotaniya et al. 2015). However, it has been realized long term irrigation with tannery effluent would result in accumulation of heavy metals in soil and groundwater resources beyond the permissible limit (Rattan et al. 2005). Among the different heavy metals, chromium (Cr) is commonly found in tannery effluent and is one of the most detrimental elements to plant growth (Dotaniya et al. 2014, 2016). Tannery effluent is the major source of influx of Cr into the biosphere. About 40% of the total Cr used in the tannery industry is released in to the environment as hazardous element (Barnhart 1997). Higher Cr in soil, furthermore affects the germination, growth of root and shoots as well as crop yield (Shanker et al. 2005).

Kanpur city in Uttar Pradesh, India is aptly called “leather city” as some of the largest tanneries in India are situated here. Over the years, soil and water pollution in the Kanpur region has drastically reduced the crop yields by 25%–40%. The total cropping area also decreased due to the pollution (Sahu et al. 2007). Similarly in a span of 20 years, the total cropped area has reduced by about 10.5% in Vellore district of India due to loss of productivity caused by the tannery effluent irrigation (Dakiky et al. 2002). The

✉ M. L. Dotaniya
mohan30682@gmail.com

[1] ICAR-Indian Institute of Soil Science, Nabi Bagh, Berasia Road, Bhopal 462 038, India

tannery effluent has high chemical oxygen demand (COD), biochemical oxygen demand (BOD), carbonate, chloride, calcium, and chromium level than the permissible limit of Bureau of Indian Standards (BIS). Therefore, most of the tannery effluents are not suitable for irrigation (Singh and Rao 2013). Undesirable constituents in wastewater from tannery can harm human health and the environment and is an issue of concern to public agencies responsible for maintaining public health and environmental quality.

There has not been a comprehensive study to assess the heavy metal concentration in the tannery effluents and the accumulation level in agricultural fields and groundwater. Therefore, the current study aimed to estimate the heavy metals accumulation in agricultural soils and groundwater of sub-urban areas of Kanpur where the tannery effluents has been used for irrigation for over 50 years. The geo-accumulation index was used as a method to assess the soil pollution in this region. The influence of soil particle size distribution and organic carbon on heavy metal accumulation was also studied to understand the behaviour of these heavy metals in these soils.

Materials and Methods

Tannery effluents, groundwater and soil samples from different agricultural fields were collected in and around tannery industrial areas of Kanpur city (26°46′ N and 80°35′ E). Kanpur experiences extremes of temperature with a minimum of 0°C in the winters while it goes up to 48°C in summers with southwestern monsoon rains in July–September. Some rainfall is also received during March–April.

Effluents were collected from twenty tannery industry sites. Soil (17 locations) and groundwater (12 locations) samples were collected from in tannery effluent irrigated areas. About six composite soil samples were collected from uncontaminated groundwater irrigated sites in the nearby areas. Groundwater levels in the region vary from 8 to 11 m. Soils were collected randomly from the effluent irrigated fields and non-effluent irrigated fields. Soil sampling was carried out from 0 to 15 cm depth using a tube auger. The samples were collected by adopting Simple Random Sampling (SRS) technique. After collection, soils were air dried under shade and stored in polythene bags. The groundwater samples were collected from the tube wells and stored in thoroughly rinsed plastic bottles.

Electrical conductivity (EC) and pH of effluent, soil and groundwater samples and texture of soils were determined as per the standard methods (Singh et al. 2005). The oxidizable organic carbon (OC) in soil was analyzed by wet oxidation method (Walkley and Black 1934). Total heavy metals viz., Zn, Cu, Pb, Ni, Cr, As and Cd were estimated by the procedure as described by Page et al. (1982). The effluent and groundwater, samples were digested with dilute nitric acid (HNO_3) (Huamain et al. 1999), and the soils were digested with aqua regia (75% concentrated HCl + 25% concentrated HNO_3). The digested materials after proper dilution with double distilled water were analyzed for heavy metals using Inductively Coupled Plasma-Optical Emission Spectrophotometer (ICP-OES; Perkin Elmer Precisely Optima 2100 DV). The detection limit for the heavy metals was 0.001 ppm.

Geo-accumulation index is widely used for assessing heavy metal contamination in sediments (Ball and Izbicki 2004; Chabukdhara and Nema 2012), dust (Kong et al. 2011); and trace metal pollution in agricultural soils (Wei and Yang 2010). The geo-accumulation index, in the present study, was calculated using the following formula described by Muller (1969).

$$I_{geo} = \log_2 \frac{C_n}{1.5B_n}$$

where, I_{geo} stands for geo accumulation index; C_n is the soil trace metal concentration (mg kg^{-1}) and B_n geochemical baseline concentration (mg kg^{-1}) i.e. the mean trace metal concentration in the soils unirrigated with tannery effluents contaminated water.

The soil sample with $I_{geo} \leq 0$ indicates unpolluted and classified under class I. Similarly, I_{geo} values 0–1, 1–2, 2–3, 3–4, 4–5 and >5 indicates unpolluted to moderate polluted (class II), moderate polluted (class III), moderate to heavily polluted (class IV), heavily polluted (class V), heavily to extremely polluted (class VI) and extremely polluted (class VII), respectively. Each parameter in the study was analyzed in triplicates. The descriptive statistical analysis for pH, EC, OC and heavy metals and correlation co-efficient of soil particle and organic carbon with heavy metals were carried out using the statistical package SAS 9.3.

Results and Discussion

The various characteristics of the tannery effluents collected from Kanpur region of India are presented in Table 1. The pH and EC of the effluent samples at source point were 8.2 and 11.53 dS m^{-1}, respectively. The high EC value in the effluents might be accumulation of salts that were used during the tanning process of leather manufacturing. The mean heavy metal content (mg L^{-1}) in the effluent samples were 0.005, 0.017, 15.83, 0.037, 0.137, 0.007 for Cd, Pb, Cr, Ni, Zn and As, respectively. The data revealed that the tannery effluent was highly polluted with higher concentration of Cr exceeding the maximum permissible limit set up by the Uttar Pradesh Pollution Control Board (UPPCB), India. Similar high concentration of Cr were observed by Gurjar and Yadav (2013) in tannery effluents in southwest Delhi.

Table 1 Physico-chemical properties of tannery effluents and groundwater collected from tannery industrial area of Kanpur

Parameters	Effluent at source point		Effluent use for irrigation (pre-treated water)		Groundwater		Maximum permissible value[a] (mg L^{-1})
	Mean ± s.d.	Range	Mean ± s.d.	Range	Mean ± s.d.	Range	
pH	8.20 ± 0.14	7.91–8.76	7.760 ± 0.06	7.66–7.97	7.78 ± 0.25	7.46–8.26	–
EC (dS m^{-1})	11.53 ± 5.14	2.35–13.15	4.076 ± 0.17	3.69–4.70	2.84 ± 0.20	2.48–3.15	–
Cd (mg L^{-1})	0.005 ± 0.00	ND-0.02	0.013 ± 0.00	0.01–0.01	ND	ND	0.01
Pb (mg L^{-1})	0.017 ± 0.01	ND-0.07	0.013 ± 0.01	ND-0.06	0.004 ± 0.00	ND-0.01	0.01
Cr (mg L^{-1})	15.83 ± 10.79	1.53–57.35	1.985 ± 0.58	0.77–4.16	0.024 ± 0.01	0.02–0.04	0.03
Ni (mg L^{-1})	0.037 ± 0.02	ND-0.12	0.016 ± 0.00	0.01–0.02	ND	ND	0.01
Zn (mg L^{-1})	0.137 ± 0.09	ND-0.48	0.184 ± 0.03	0.08–0.28	0.126 ± 0.05	0.16–0.18	0.30
As (mg L^{-1})	0.007 ± 0.01	ND-0.03	0.001 ± 0.00	ND-0.00	ND	ND	–

ND not detected

[a]Maximum permissible concentration as defined by USEPA (2009) for drinking purpose

This might be due to use of large amount of chromium sulphate salts for tanning processes in the industry. The significant amount of heavy metals other than Cr might have originated from other industrial effluents that were also discharged into the same sewage channel (Gurjar and Yadav 2013). Contaminated water used in the fields for irrigation after pre-treatment had lower pH (7.8) and EC (4.1 dS m^{-1}) when compared to the corresponding values of the effluent at source (not treated). The total concentration of heavy metals, particularly Cr had decreased after the treatment of the effluent and the mean content was found to be 1.99 mg Cr L^{-1}as compared to mean value of 15.83 mg Cr L^{-1} in the untreated effluent sample (Table 1).

The groundwater samples were also collected in 2013 from the area where these effluents were used for irrigation. The data revealed that the pH of groundwater samples varied from 7.46 to 8.26 and EC from 2.48 to 3.15 dS m^{-1}. The mean total heavy metal content in groundwater for Pb, Cr and Zn was 0.004, 0.024 and 0.126 mg L^{-1}, respectively (Table 1). The heavy metal concentration in majority of the groundwater samples were within the safe limit as per USEPA (2009), while only two samples out of 12 samples showed Cr concentration (0.040 and 0.034 mg L^{-1}) exceeding the safe permissible limit suggesting that there is chance for contamination of groundwater due to surface irrigation with tannery effluent contaminated water. Leaching of heavy metals from soil probably increased the groundwater heavy metal concentration (Ball and Izbicki 2004). Moreover, the water table is very shallow (not more than 4–6 meters depth) in Indo- Gangatic Plains (Rattan et al. 2005). Among the various industrial effluents, the tannery effluent was shown to be the most dangerous pollutant that affected the quality of water bodies (Parithabhanu and Khusnumabegam 2013). Further, the small scale tannery industries discharge the effluent directly into the environment without any pretreatment and thus pose greater threat to surface water bodies (Sharmila et al. 2013). The hexavalent Cr is of greater concern due to its great toxicity to crop plants and also known to be carcinogenic and mutagenic to living organisms (Nakano et al. 2001). Singh et al. (2001) reported high levels of Cr in groundwater samples in carpet industrial area of eastern Uttar Pradesh, indicating the leaching of metal released from the industry. It has been shown that Cr(VI) has high mobility than Cr(III) and that the latter is preferentially retained by the soil.

In the agricultural fields where tube well water was used for irrigation purpose, the pH of soil varied from 7.21 to 7.33 and concentrations (mg kg^{-1}) of Cu (33.1–72.3), Cd (0.6–0.80), Pb (5.9–11.60), Cr (15.2–33.2), Ni (7.9–14.2), Zn (28.5–99.3) and As (3.1–5.1) was observed in the soils (Table 2). The pH of the soil was found to be in optimum range and also had the lowest concentrations of heavy metals. This concentration did not show any detrimental effect on plant growth and yield (Gyawali and Lekhak 2006). On the other hand, soil samples collected from agricultural field where contaminated water (tannery effluent) was used for irrigation purpose had pH 7.15–8.33, EC (dS m^{-1}) 1.23–3.56 and OC (%) 0.72–1.52. The total heavy metals content was higher in tannery effluent irrigated soil than that of groundwater irrigated soils (Table 2).

Thus the results showed that the use of contaminated water for irrigation resulted in the buildup of the heavy metals in the crop field. It was observed that continuous use of tannery effluent for vegetable and wheat production had resulted in building up 28–30 times more Cr in the soils than the soils irrigated with groundwater. Moreover Sinha et al. (2006) reported high Cr accumulation in soils of tannery irrigated areas of Kanpur. Chromium concentration in soil increased approximately by 16%–30% (Dhungana and Yadav 2009) and other heavy metals by 2–3 times (McGrath et al. 1994), when tannery effluent was used for irrigation. The higher buildup of salt and metal

Table 2 Physico-chemical properties and heavy metals concentrations of tannery effluents and ground water irrigated soil samples

Parameters	Ground water irrigated soil		Tannery effluent irrigated soil	
	Mean ± s.d.	Range	Mean ± s.d.	Range
pH (1:2.5)	7.27 ± 0.02	7.21–7.33	7.65 ± 0.08	7.15–8.33
EC (dS m^{-1})	0.37 ± 0.06	0.19–0.56	1.75 ± 0.16	1.23–3.56
Sand (%)	53.42 ± 4.40	49.2–61.5	52.94 ± 3.68	48.2–61.5
Silt (%)	32.32 ± 5.51	24.4–36.8	32.96 ± 4.10	26.3–38.4
Clay (%)	14.26 ± 2.73	12.0–19.4	14.10 ± 2.93	8.2–19.7
OC (%)	0.42 ± 0.06	0.33–0.68	1.08 ± 0.06	0.72–1.52
Cu (mg kg^{-1})	50.00 ± 6.46	33.10–72.30	54.15 ± 3.55	35.30–82.80
Cd (mg kg^{-1})	0.70 ± 0.03	0.60–0.80	5.19 ± 0.85	2.30–14.10
Pb (mg kg^{-1})	7.94 ± 0.97	5.90–11.60	37.73 ± 2.45	23.70–58.80
Cr (mg kg^{-1})	23.50 ± 3.47	15.20–33.20	505.88 ± 54.11	252.40–971.70
Ni (mg kg^{-1})	11.14 ± 1.03	7.90–14.20	25.91 ± 0.52	22.90–30.30
Zn (mg kg^{-1})	50.40 ± 12.62	28.50–99.30	210.55 ± 14.07	138.70–338.10
As (mg kg^{-1})	3.84 ± 0.37	3.10–5.10	8.71 ± 0.31	6.80–11.00

concentration in agricultural field deteriorates the soil quality and affects the sustainability (Singh et al. 2004).

The correlation studies relating soil organic carbon and particle size distribution with heavy metals showed that organic matter and clay fractions were significantly correlated ($p = 0.05$) with heavy metal accumulation and distribution in the effluent irrigated soils (Table 3). On the other hand, there were no correlations ($p = 0.05$) observed in uncontaminated groundwater irrigated soils. Also, it was found that clay correlated with positively charged metals like Cu, Cd, Ni, Zn, and Pb, but did not show any relationship with negatively charged anions i.e., As and Cr. However organic carbon showed significant positive relationship with all the metals except As. The ion exchange and adsorption properties of the clay and organic matter might have influenced the distribution and accumulation of these heavy metals. Sand particles were found to have negative relationship with heavy metal contents. The similar kinds of results were reported by many workers (Alloway 1995; Naidu and Harter 1998; Alamgir et al. 2015). Sherene (2010) concluded that the fine textured soils contain higher amounts of Pb (3889 mg kg^{-1}) and coarse textured soil contains (530 mg kg^{-1}) lower amount of Pb. Similarly Jeyabaskaran and Sree Ramulu (1996) concluded that the content of DTPA-extractable metals in light textured soils were low in soils irrigated with sewage water for the past 50 years. Disla et al. (2008) also confirmed the importance of soil properties such as texture and organic matter or carbonates on the behavior of heavy metals through multiple regression analyses. Due to high specific surface area organic matter can form complexes with heavy metal and consequently influence their distribution. Further McLean and Bledsoe (1992) found that adsorption of metal cations has been correlated with such soil properties as pH, redox potential, clay, soil organic matter, Fe and Mn oxides, and calcium carbonate content.

The increasing level of heavy metal contamination in agricultural field led to higher geo-accumulation index (I_{geo}) value (Table 4). The I_{geo} of all the heavy metals in the present study had shown values greater than 0 indicating that the soils were contaminated with heavy metals, but the extent of contamination varied with the type of heavy metals. For instance, the I_{geo} of Cu, Pb, Ni, Zn and As was found to be in the range of 0–1 representing the level of

Table 3 Correlation Co-efficient of particle size distribution and organic carbon with heavy metals

Heavy metal	Tannery effluent irrigated soil				Ground water irrigated soil			
	Sand	Silt	Clay	OC	Sand	Silt	Clay	OC
Cu	−0.043	−0.303	0.488*	0.558*	−0.031	0.095	−0.142	−0.331
Cd	−0.020	−0.355	0.519*	0.474*	−0.134	−0.080	0.378	0.136
Pb	−0.143	−0.238	0.522*	0.639*	0.929	−0.760	0.039	0.893
Cr	−0.150	−0.086	0.306	0.507*	−0.142	0.182	−0.137	−0.074
Ni	−0.161	−0.207	0.498*	0.540*	0.824	−0.610	−0.096	0.635
Zn	−0.155	−0.178	0.456*	0.595*	−0.300	0.080	0.322	0.057
As	−0.028	−0.291	0.435	0.354	−0.300	−0.751	−0.005	0.756

*Significant at 5%

Table 4 Geo-accumulation index of heavy metals in tannery effluent irrigated soils of Kanpur

Elements	Value	Class	Description
Cu	0.22	II	Unpolluted to moderate polluted
Cd	1.49	III	Moderate polluted
Pb	0.95	II	Unpolluted to moderate polluted
Cr	4.32	VI	Heavily to extremely polluted
Ni	0.47	II	Unpolluted to moderate polluted
Zn	0.84	II	Unpolluted to moderate polluted
As	0.46	II	Unpolluted to moderate polluted

contamination to be between unpolluted to moderate level by these metals. However, in case of Cd and Cr, the I_{geo} values were 1.49 and 4.32, respectively; which showed moderate contamination for Cd and extreme level of contamination for Cr. Use of tannery effluent for irrigation might have increased the significant amount of Cr build up in soil (Gyawali and Lekhak 2006). The significant amount of other metals, particularly Cd might have originated from other wastewater discharged into the tannery effluent carrying channels.

The study revealed that the use of tannery effluent for crop production has led to built up of Cr in the soil by 28–30 times. The change in the geo-accumulation index of heavy metals from class II (unpolluted to moderate polluted) to VI (heavily to extremely polluted) in the study area is a sign of heavy metal pollution in agricultural field. The increased pollution level may adversely affect the soil health and likely to contaminate food chain. Therefore, the government should take initiatives to strictly monitor the effluent treatment plants regularly and also spread the awareness among the public on impact of tannery effluents on soil–plant–human systems through government and nongovernmental organizations (NGOs) in the tannery effluent irrigated areas. Further, use of mixed industrial effluents for agricultural activities should be avoided and more investigation is needed to study the risk of multi metal toxicity.

Acknowledgements Authors are highly thankful to supporting staff of the Division of Environmental Soil Science, ICAR-Indian Institute of Soil Science, Bhopal for necessary help during the study. Authors are also thankful to Dr. S. R. Mohanty for valuable comments. This study is a part of the project "Tannery constituent interaction effect on spinach" and funded by ICAR-Indian Institute of Soil Science, Bhopal (Grant No. IXX07989).

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