

STRATEGIES FOR THE REMEDIATION OF CADMIUM AND CHROMIUM FROM INDUSTRIAL EFFLUENTS IN RESPONSE TO *Amaranthus cruentus*, *Spinacia oleracea* and *Amaranthus viridis* OF BANGLADESH

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Abstract

Discharge of industrial effluents and their remediation in relation to crop production are the major concerns. Accordingly, the color, pH, EC, total dissolved solids (TDS), dissolved oxygen (DO), chemical oxygen demand (COD), total hardness, chloride, CO_3 , HCO_3 , alkalinity, Cu, Cd, Pb, Mn, Zn, and Cr contents in the effluents discharged from Hazaribagh tannery and Tejgaon textile industries in Dhaka were determined. These effluents had no significant ($p \leq 0.05$) effects on soil pH but exerted significant positive effects on the CEC of the soil. The TDS of the effluents were also high but it decreased by alum [$\text{K}_2\text{SO}_4 \text{ Al}_2(\text{SO}_4)_3 \cdot 24 \text{ H}_2\text{O}$] treatment (0.1%). The DO of the effluents was low and the COD was high leading to a serious threat for aquatic lives. The concentrations of Cd, Pb, Zn, Mn and Cr were high. Filtration through natural sand-gravity-filter acts like Effluent Treatment Plant-ETP and coagulation of effluents by alum decreased the concentrations of Cd and Cr below the permissible limits. The treated effluents were used for the production of vegetables of red amaranth (*Amaranthus cruentus*), green spinach (*Spinacia oleracea*) and green amaranth (*Amaranthus viridis*) grown in a non-polluted soil under pot experiments. Application of treated effluents was found to have significant ($p \leq 0.05$) positive effects on the biomass production of the vegetables. The highest amounts of assimilation of Cd in plants were recorded for the Green spinach followed by Green amaranth and for Cr, the order was green spinach followed by red amaranth and green amaranth vegetables grown under the treated effluents of tannery and textile industries. The present study revealed that the natural sand-gravity-filter (i.e., Economic ETP) and alum treatments were found to be effective not only for the remediation of polluted effluents but also improved the growth of vegetables. The use of natural filter and/or alum treatment can be practiced for the remediation of pollution of industrial effluents before discharging from industries.

Key words: Natural sand-gravity-filter; Phytoremediation; Purification; Industrial effluents; Heavy metals.

INTRODUCTION

The problem of industrial effluent has increased significantly in Bangladesh as a consequence of rapid industrialization in the post-independence period, prior to which our economy was largely agrarian. Contamination of the environment by heavy metals has increased abruptly at the beginning of the 20th century, as a result of the industrial revolution and excessive population growth, posing major environmental and human health problems worldwide (Abdehfez and Li 2014). These metals cannot be easily degraded to harmless products and the cleanup usually requires their removal (Wu *et al.* 2018). Several remediation technologies are available for treating heavy metals contaminated soils. Of them, phytoremediation has attracted increasing attention and is a promising technology for addressing soil contamination problems. It is a cost-effective green technology based on the use of plants to remove, metabolize, assimilate, or adsorb hazardous materials in soil (Wu *et al.* 2015). Phytoremediation involves several specific subsets, of which phytoextraction and phytodegradation are currently the most developed for the phytoremediation of metals and organics in contaminated soil (Lee 2013, Mani *et al.* 2015). Phytoextraction is the process of using plants, especially hyper-accumulator plants, to extract or concentrate metals or organics to harvestable biomass, whereas phytodegradation is the process of using the root exudates of plants to stimulate microbial activity whereby the enhancement of mineralization at the root-soil interface is attributed to microorganisms

associated with the root surface (Mani *et al.* 2015). The industrial sector is gradually playing a more important role in Bangladesh's economy. However, this sector has altered the fragile environment of the surrounding area. A huge amount of industrial effluents are generally discharged into surface water courses, sewers and on the land, without treatment or inadequately treated. This has created a problem of surface and subsoil water pollution and soil pollution. Industrial effluents contain some heavy metals such as Pb, Cd, Zn, Cu and Cr *etc.* Once a soil is contaminated by metals, it remains so indefinitely. The fate of heavy metals added to soils, including their mobility and reactions on the soil and their subsequent uptake and distribution in plants is, therefore, of critical importance in relation to human health.

The Department of Environment (DoE) in Bangladesh has identified 900 polluting industrial units, most of which do not have treatment facilities for effluent and wastes. Department of Environment (DoE) and Ministry of Industries (MoI) with the assistance of Asian Development Bank (ADB) carried out a study on pollution aspects in Bangladesh, which conclude the ranking of the major polluting industry sector as (i) Paper and pulp, (ii) Textile (dyeing and printing) and (iii) Tanneries. These industrial sectors are major pollutants due to the high discharge level of organic materials and chemicals, which are hazardous to environment. Heavy metals even in trace amounts destroy enzymes of living cells (Rahman 1992). As a consequence of anthropogenic activities, there is a progressive increase in soil and water pollution by heavy metals. These metals can be remediated by growing plants or vegetables, i.e., phytoremediation. On the other hand, industrial effluents can be treated by natural sand-gravity-filter (Economic ETP) and chemically. The treated effluents can be discharged into surface watercourses, sewers and on the land, so the problem of pollution can be minimized, if not totally eliminated. This practice will not only be beneficial for our soil but would also be a constructive step toward minimizing pollution. Nevertheless, it has to be ensured that the treated effluent is safe enough to be used for irrigation purposes. The effluent concentrations of Cd, Cr, and their mobility and responses to plants are desired concerns. Environmentally friendly remediation technologies for cleanup of Cd and Cr contaminated areas have received increasing interest. Accordingly, the objectives of this research were: (i) Assessment of the effluents generally discharge in the environment, (ii) Find out the suitable strategies for the reclamation, phytoremediation and purification of the industrial effluent, and (iii) Aware of people in various government, non-government and research organization regarding use, recycling and treatment processes of effluents (sand-gravity-filter - Economic ETP) with minimum cost as well as reuse of the huge amounts of effluents for agricultural purposes.

MATERIAL AND METHODS

Sampling sites

Industrial sectors are major pollutants due to the high discharge level of organic materials and chemicals, which are hazardous to environment. Accordingly, Hazaribagh Tannery and Tejgaon Textile Industrial areas were considered for this study. Hazaribagh tannery units generated 8.47 million liter liquid wastes and 98 MT solid wastes every day, which have not only been poisoning the soil, water and air round but also poisoning the health, houses and utensils of those soil-plant ecosystems. The Tejgaon industrial area in the outskirts of Dhaka city is the only exclusive industrial region and it is an area of various industrial units. The wastes from the textile industries are discharged into the low-lying swamps and 'Canals' in the adjacent area near Badda of Dhaka city. Only a few of the industrial units have technical effluent treatment plants. Some of the industrial

wastes also find their way into the Gulshan Lake (eastern) of Dhaka city. The effluents were being discharged via gravity-flow channel systems. Concrete drains, cement-asbestos and concrete/PVC-pipes were being used for carrying the discharge to the low-lying areas or the adjacent swamps and marshes, which have now turned into ideal breeding grounds for mosquitoes - a serious problem in the present days. In the developed countries, the wastes and effluents are treated to safe level before their discharge into the natural ecosystems. In Bangladesh, these are disposed of at random into soils, canals and rivers without any treatment.

The effects of effluents on soil-plant-water qualities were evaluated through simulation studies in the laboratory and pot experiments under polyethylene house at the premises of the Department of Soil, Water and Environment in the University of Dhaka. Considering the cluster of industries, composite samples from each site were collected from the distances of about 50-100 meters far away from the factories. Moreover, the color, acidity, alkalinity levels were varied even within an hour. The composite effluent from each site was examined for possible reclamation measures. The color, pH, EC, TDS, DO, COD, total hardness, chloride, CO_3^{2-} , HCO_3^- , alkalinity, Na, K, S, Cu, Cd, Pd, Mn, Zn, and Cr in the effluents and the responses of selected heavy metals to crops and soils under different treatments were evaluated through phytoremediation and water purification measures following standard methods. Filtration of effluents through natural sand-gravity-filter (Effluent Treatment Plant-ETP) and coagulation by alum (0.1%) treatments were used for the production of vegetables, viz. red amaranthus (*Amaranthus cruentus*), green spinach (*Spinacia oleracea*) and green amaranth (*Amaranthus viridis*) grown in a non-polluted silt loam soil (Table 1) collected from Dhamrai series was used for this study under pot experiment.

Table 1. Some physico-chemical properties of the soil used (0-15 cm depth).

Properties	Values
pH (Jackson 1973)	7.5
EC ($\mu\text{S}/\text{cm}$; Richards 1954)	200
TDS (mg/L)	100
Moisture (%; Black 1965)	1.5
Textural class (Piper 1966)	Silt loam
Organic carbon (%; Nelson and Sommers 1982)	0.37
CEC (c mol/kg; Black 1965)	5.5
Exchangeable Ca (c mol/kg; AAS*)	4.7
Exchangeable K (c mol/kg; Flame photometer)	0.13
Exchangeable Na (c mol/kg; Flame photometer)	0.12
Available N (mg/kg; Jackson 1973)	29
Available P (mg/kg; Olsen <i>et al.</i> 1954)	1.6
Available K (mg/kg; Pratt 1965)	55
Available S (mg/kg; Sakai 1978)	0.50
Cd (mg/kg; AAS)	0.12
Cr (mg/kg; AAS)	29.0

*AAS = Atomic Absorption Spectrophotometer

The effects of purification and phytoremediation strategies of the heavy metals through the treatments of industrial effluents on the vegetables and on soil properties were evaluated at three stages 30, 60 and 90 days after seed sowing of growth having three replications. The plant shoots, and roots of red amaranthus (*Amarunthus cruentus*), green spinach (*Spinacio oleracea*) and green amaranth (*Amarunthus viridis*) were analyzed for Cd and Cr contents in these vegetables. Statistical analyses were done following MS Office package.

RESULTS AND DISCUSSION

The pHs of the original effluents collected from the tannery and textile effluents were 7.4 and 7.9, respectively (Table 2). The pH values of the filtrate and chemically treated effluents of both the industries were increased by 0.6 and 0.3 units, respectively. The EC value of the original tannery, filtrate and chemically treated effluents were 11620, 9680 and 720 $\mu\text{S}/\text{cm}$, respectively. The EC values of original, filtrate and chemically treated textile effluents were 1520, 1230 and 1280 $\mu\text{S}/\text{cm}$, respectively (Table 2). Values exceeding 4000 μS are the harmful limit for rice (Ponnamperuma 1985). But the EC value of 720 $\mu\text{S}/\text{cm}$ as obtained in the chemically treated tannery effluent was below the harmful limit. This indicates that the filtration and/or chemical treatment for the effluents were effective to reduce the salinity (EC) of the effluents. The total dissolved solids of the studied samples were also high (5660 mg/L) around the tannery factory site. However, the TDS of the chemically treated tannery effluents was found to be decreased up to the permissible limit as because of the use of coagulant (alum), which decreased the density of effluents resulting low yields of TDS.

Table 2. Some selected properties of Tannery and Textile effluents.

Parameter	TnEfo	TnEfF	TnEfc	TtEfo	TtEfF	TtEfc
Color	Yellowish blue	Light yellow	Light yellow	Bluish black	Blue	Blue
pH	7.4	8.2	8.0	7.9	8.2	8.2
EC ($\mu\text{S}/\text{cm}$)	11620	9680	720	1520	1230	1280
TDS (mg/L)	5660	4500	590	900	730	760
DO (mg/L)	Trace*	Trace	Trace	1.7	2.3	2.0
COD (mg/L)	300	250	250	60.0	50.0	45.0
Total hardness (mg/L)	1800	2600	1950	690	430	400
Chloride (mg/L)	2.25	2.11	1.93	0.23	0.20	0.19
CO_3^{2-} (g/L)	-	-	-	0.22	-	-
HCO_3^- (g/L)	0.61	3.0	0.52	0.85	0.43	0.21
Alkalinity (g/L)	0.61	3.0	0.52	1.07	0.43	0.21
Na (mg/L)	72.5	66.5	66.0	37.5	31.0	35.5
K (mg/L)	17.5	20.75	20.75	12.0	3.75	3.75
S (mg/L)	206.83	103.88	424.23	88.10	105.34	153.0
Cu (mg/L)	0.04	0.22	0.13	0.13	0.18	0.00
Cd (mg/L)	0.09	0.00	0.00	0.01	0.00	0.00
Pb (mg/L)	0.4	0.02	0.01	1.95	0.08	0.06
Cr (mg/L)	400	260	169	255	147	71

TnEfo = Tannery effluent original, TnEfF = Tannery effluent filtered, TnEfc = Chemically treated tannery effluent, TtEfo = Textile effluent original, TtEfF = Textile effluent filtered, TtEfc = Chemically treated textile effluent. *Trace = Not within the detectable range.

The dissolved oxygen (DO) of the tannery effluents was very low, i.e., undetectable range, indicating that this effluent is harmful for aquatic lives. The DO was very low in the textile effluents too leading to a serious threat for the same. The chemical oxygen demand (COD) of the tannery effluents was 300 mg/L, which is above the permissible limit of inland surface water. The COD of the textile original, filtrate and chemically treated effluents were 60, 50, 45 mg/L, respectively which were also below the permissible quantity. The total hardness of the tannery original, filtrate and chemically treated effluents were 1800, 2600 and 1950 mg/L, respectively which are very high. The total hardness of the textile original, filtrate and chemically treated effluents were also high and the values were 690, 430 and 400 mg/L, respectively. However, the value of the textile filtrate and chemically treated effluents were 430 and 400 mg/L, respectively which remained below the permissible limit. The chloride concentration of the tannery original, filtrate and chemically treated effluents were found above the permissible limit. But for the textile original, filtrate and chemically

treated effluents, the concentrations of chloride were determined near to or below the permissible limit.

Alkalinity of water is generally measured through the determination of the amounts of its content of carbonate (CO_3^{2-}) and bicarbonate (HCO_3^-). Carbonate concentrations were not detected in the tannery original, filtrate and chemically treated effluents. The carbonate concentration was found 0.22 g/L in the textile original effluents but its concentration was not detected in textile filtrate and chemically treated effluents. The concentrations of HCO_3^- were 0.61, 3.0, and 0.52 g/L in the tannery original, filtrate and chemically treated effluents and for the textile industries, the values were 0.85, 0.43 and 0.21 g/L for original, filtrate and chemically treated effluents, respectively. Sodium concentrations were determined 72.5, 66.5 and 66 mg/L, respectively in the tannery original, filtrate and chemically treated effluents. On the other hand, its concentration was quite low as 37.5, 31.0 35.5 mg/L, respectively in the textile original, filtrate and chemically treated effluents, which might be attributed to the variation in use of sodium containing chemicals such as urea, soap, etc. in textile industry.

The high content of S (206.83 mg/L) was determined in tannery effluents. The content of sulfur at 0.5 to 1.0 mg/L level is fatal to sensitive fish (trout) even in neutral or alkaline water. Similarly, the high content of S (88.1 mg/L) was also found in textile effluents. And S (153.0 mg/L) was found in chemically treated textile effluents. The high content of S affects life and also interferes with the biological treatment processes. The concentrations of sulfur and nitrogen were also in hazardous levels at Hazaribagh tannery area.

Among the heavy metals Cu, Cd, Pb, Zn, Mn, and Cr contents were determined high in effluents of Hazaribagh Tannery. The high concentrations of metal particularly chromium in the effluents in the tannery effluents were the result of discharge of the metals from the various leather treatment processes. The process of filtration and coagulation (using chemical agents) of the effluents were found to be decreased the concentrations of those elements below the permissible limits.

Cadmium content

Tannery industries have discharged not only high amount of Cd through the effluents but also lasting significant amounts of Cd in the soil-plant-water systems. In a few cases, Cd was also recorded above the toxic levels in the soil used (Fig. 1). There were significant ($p \leq 0.05$) effects of effluents (original, filtrate and chemically treated tannery and textile effluents) treatments regarding to the growth of vegetables of red amaranthus (*Amaranthus cruentus*), green spinach (*Spinacia oleracea*) and green amaranth (*Amaranthus viridis*). Application of treated effluents was found to have significant positive effects on the biomass production of the vegetables. Results showed that the green spinach (palong shak) was found to be assimilated the highest amounts of Cd (25 mg/kg) in shoots as compared with the shoots of green amaranth (data shak) and red amaranthus (lal shak) as determined for the filtrate tannery effluents treated vegetables (Fig. 2).

There were significant ($p \leq 0.05$) effects of effluents (original, filtrate and chemically treated tannery and textile effluents) treatments regarding to the growth of vegetables (red amaranthus, green spinach and green amaranth). Application of treated effluents was found to have significant positive effects on the biomass production of the vegetables. Results showed that the spinach (palong shak) was found to be assimilated the highest amounts of Cd (25 mg/kg) in shoots as compared with the shoots of data and lal shak as determined for the filtrate tannery effluents treated vegetables (Fig. 2). But in the filtrate textile effluents treatment, the variety of red amaranthus (lal shak) was recorded for

the highest uptake of Cd (12.5 mg/kg) in the roots as compared the roots of green spinach (palong shak) and green amaranth (data shak) (Fig. 3).

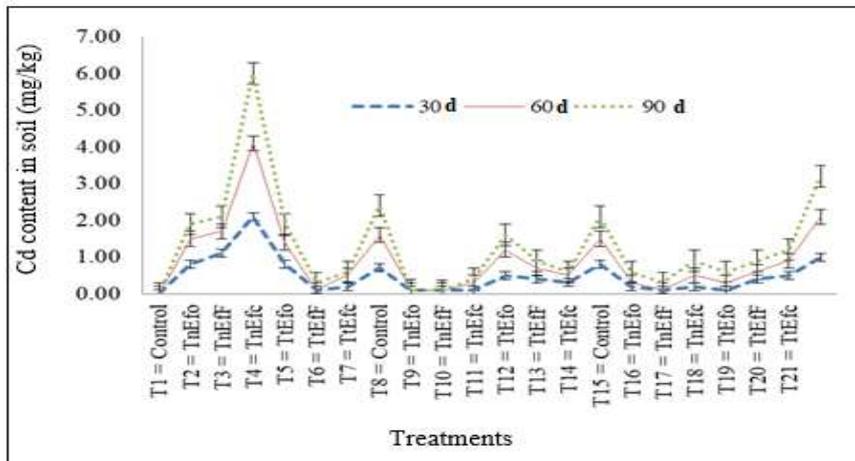


Fig. 1. Cadmium content in soil at different days (d) of harvesting of vegetables as influenced by various treatments (T₁-T₇: lal shak; T₈-T₁₄: palong shak; and T₁₅-T₂₁: data shak) of industrial effluents.

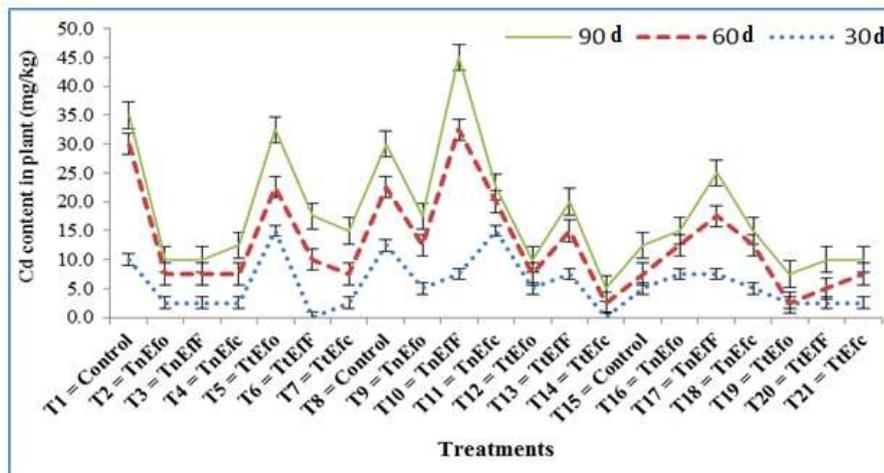


Fig. 2. Cadmium content in plant tissues at different days (d) of harvesting of vegetables as influenced by various treatments (T₁-T₇: lal shak; T₈-T₁₄: palong shak; and T₁₅-T₂₁: data shak) of industrial effluents.

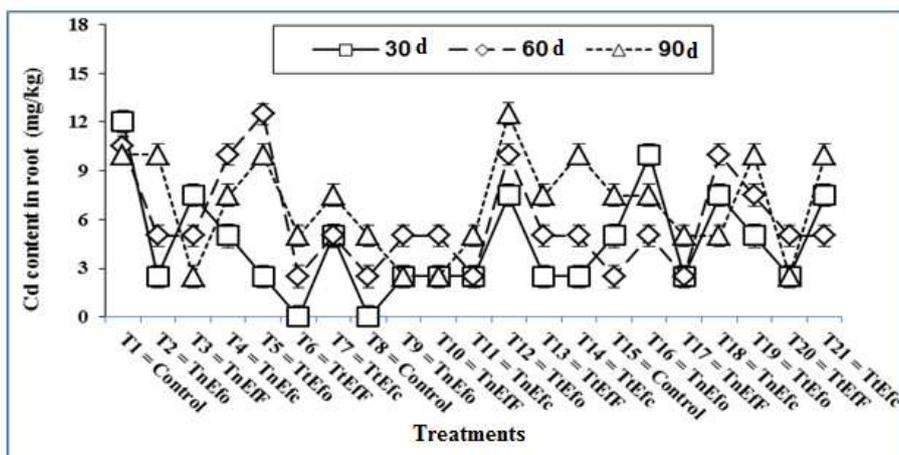


Fig. 3. Cadmium content in root at different days (d) of harvesting of vegetables as influenced by various treatments (T₁-T₇: lal shak; T₈-T₁₄: palong shak; and T₁₅-T₂₁: data shak) of industrial effluents.

It should also be mentioned that the green spinach (palong shak) was found to have assimilated the highest amounts of Cd (25 mg/kg) in shoots as compared with the shoots of green amaranth (data shak) and red amaranth (lal shak) as recorded for the filtrate tannery effluents treated vegetables. But in the filtrate textile effluents treatment, the variety of Lal shak was obtained the highest content of Cd (12.5 mg/kg) in the roots as compared with the roots of palong shak and data shak.

Chromium content

The maximum amounts of elemental Cr at all sampling times were evaluated in the soils having green spinach. The soils under red amaranth and green amaranth were also showed the similar trends of effects but the contents of Cr were higher than those of the soils under Green spinach cultivation (Fig. 4). Chemical treatment of tannery effluent by alum ranked first regarding to the reduction of Cr contents in soil at all sampling times followed by the filtration treatment and these trends were also noticed for textile effluents (Fig. 4).

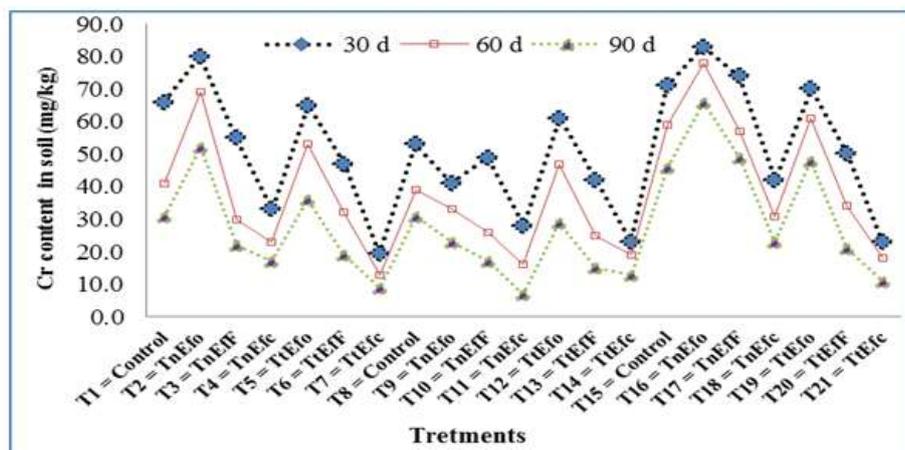


Fig. 4. Chromium content in soil at different days (d) of harvesting of vegetables as influenced by various treatments (T₁-T₇: lal shak; T₈-T₁₄: palong shak; and T₁₅-T₂₁: data shak) of industrial effluents.

The contents of the elemental Cr were determined maximum in the plant tissues of Green spinach followed by Red amaranth and Green amaranth at all sampling stages of the growth of the vegetables (Fig. 5).

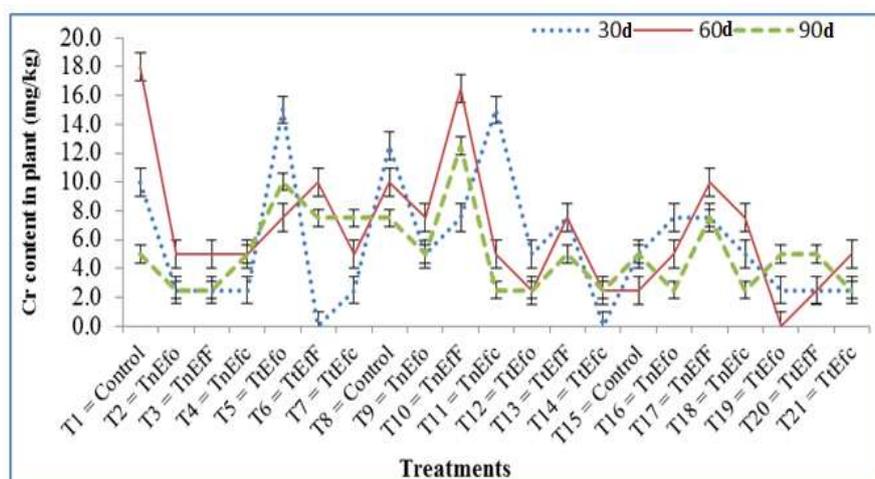


Fig. 5. Chromium content in plant tissues at different days (d) of harvesting of vegetables as influenced by various treatments (T₁-T₇: lal shak; T₈-T₁₄: palong shak; and T₁₅-T₂₁: data shak) of industrial effluents.

The contents of Cr were found to be decreased with the advent of time and the chemical treatments strikingly decreased the contents of Cr in plant tissues followed by the filtration treatment. Almost similar trends of effects of the treatments were evaluated for the contents of Cr in roots (Fig. 6). But the Cr contents were found to be increased with the advent of time of plant growth and the effects were more pronounced for red amaranthus followed by green spinach and green amaranth (Fig. 6). It should also be mentioned that the high accumulation of Cd grasses, vegetables and rice samples might threaten not only the grazing animal but also for the human beings. The contents of Cd in the post-harvest soils were well reflected by the contents of Cd in the plant tissues and clearly indicated that the vegetables accumulated high amounts of Cd which are in the toxic levels. This is dangerous for all lives because the studied vegetables are very much popular to the surrounding peoples and continuous accumulation of these heavy metals into the animal bodies will not only make sudden breakthrough of normal food chain but also may develop serious diseases like cancer.

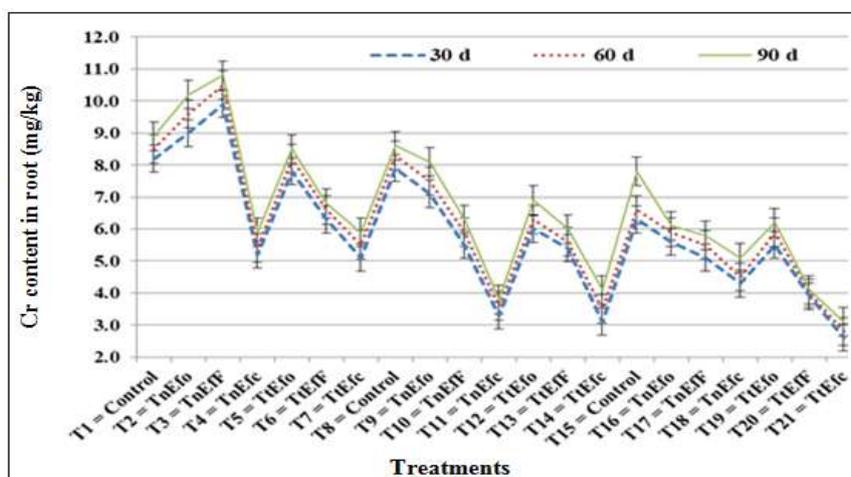


Fig. 6. Chromium content in root at different days (d) of harvesting of vegetables as influenced by various treatments (T₁-T₇: lal shak; T₈-T₁₄: palong shak; and T₁₅-T₂₁: data shak) of industrial effluents.

The present results demonstrated that the effluents used from tannery and textile industries did not affect the soil pH distinctly (Table 3). The variations in pH values were about 0.3 units in both textile and tannery effluents treated soils. The EC values of tannery effluents treated post-harvest soils were higher than that of the control treatment (Table 3). The EC value was found to be decreased remarkably after the use of natural filter. The application of chemically treated tannery effluents was found to be more effective to reduce the TDS of soils as compared to filtration practice.

Table 3. Selected properties of post-harvest soils as influenced by different industrial effluents.

Parameters	Post-harvest soils						
	Control	TnEfo	TnEff	TnEfc	TtEfo	TtEff	TtEfc
pH	7.3	7.4	7.6	7.5	7.2	7.3	7.5
EC (μS/cm)	100	200	100	200	100	100	Nd*
TDS (mg/L)	70	110	80	90	70	40	40

nd = not detected

The present study revealed that the natural sand-gravity-filter and chemical treatment through alum were found to be effective not only for the remediation of polluted soil and water but also improved the growth and quality of vegetables through the removal of toxic metals from the

effluents. The use of natural filter and/or chemical (alum, *etc.*) treatment should be practiced for the remediation of pollution of industrial effluents before discharging these effluents to surface watercourses, sewers and on the land, so that the problem of pollution can be minimized, if not totally eliminated. The practices of these processes will not only be beneficial for the polluted soils, soil-plant-water ecosystems but would also be a sustainable step towards minimizing environmental pollution. Nevertheless, it has to be ensured that the treated effluent is safe enough to be used for irrigation purposes and for this, further researches are needed.

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