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ORIGINAL ARTICLE **OPEN ACCESS**

Seasonal Variation in Chemical characteristics of Soil and Accumulation of Heavy Metals in Vegetables at Wastewater Irrigated sites at Jaipur, India

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ABSTRACT
Effect of wastewater irrigation was assessed at two agricultural sites at Jaipur city, the capital of Rajasthan, India. Variation in soil chemical characteristics and accumulation of heavy metals in two vegetables (Spinacia oleracea (spinach), Solanum lycopersicum (tomato) were studied during the pre monsoon and post monsoon period. For comparison, a control site was also studied where bore well water was used for irrigation The results indicated an increase in nutrient content in soil(OC%,NO3-N,available P and K) along with accumulation of heavy metals (Cr,Pb,Cd and Ni) in soil and vegetables. The concentration of Ni and Pb was under the permissible limits whereas Cr and Cd were found to be above the permissible limits. This concentration of metals is high enough to be accumulated in the crops grown in such soils. Higher concentration of metals was found in spinach as compared to tomato. Seasonal variation was also observed in the metal concentration in the soil and vegetables. The concentration of metals was found to be high during pre monsoon period as compared to the post monsoon period.
Keywords: *Solanum lycopersicum*, *Spinacia oleracea*, *Heavy metal*

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INTRODUCTION

The arid and semiarid regions of Rajasthan face severe problem of freshwater scarcity due to very low rainfall. This problem is further aggravated due to indiscriminate use and uncontrolled deterioration of the quality of the surface and groundwater. In addition to this growing population exerts pressure on the agriculture for production of sufficient food material. This forces the farmers to use wastewater for agriculture in the lack of fresh water. Several researches in various countries have found that municipal wastewater use in agriculture can have beneficial effects on the soil health due to its high nutrient content including organic matter, nitrogen and other elements. The effect of wastewater utilization is detrimental when the wastewater contains industrial effluents which contribute to several toxicants including heavy metals in the soil and food material. The heavy metals may also accumulate in the soil at toxic levels as a result of long-term application of untreated wastewaters [10].Heavy metals like Cd, Pb, Ni and Cr are known to cause several diseases in human beings including certain cancers. Due to their chronic toxic effect and long half life, they have potential hazardous effect on soil characteristics and human health. Thus the present study was conducted at Jaipur the pink city of Rajasthan with an aim to evaluate adverse effects of the wastewater utilization on the soil characteristics and quality of vegetables with respect of heavy metal accumulation.

MATERIALS AND METHOD

Study area: The study was conducted at two wastewater irrigated agricultural fields at Jaipur viz; Mansarovar and Pratap Nagar. For comparison, an agricultural field using bore well water for irrigation was also studies as the control site. Seasonal vegetables are grown at the study sites using wastewater from the *Amanish nala*, a common drainage passing from near the agricultural fields. The *nala* collects wastewater of several industries as well as municipal effluent while running approximately 35 km through in the city. While passing through the printing units situated at Sanganer area of the city, the wastewater is contaminated with several heavy metals emitted from the untreated effluent due to the use of chemical dyes in these units. This *nala* passes through our study sites as well from where farmers are using wastewater for growing seasonal vegetables.

Sampling: The soil samples were collected for pre-monsoon period (December 2011- June 2012) and post monsoon period (August –November 2012).

Soil samples from the wastewater irrigated and control sites were collected twice a month. It was taken care that the irrigation with wastewater has been done 3-4 days prior to sampling. The soil samples were collected from a depth of 0-15 cm from different places of the field. The samples were then kept in the duly marked polyethylene bags and taken to the laboratory for analysis. The soil was air dried prior to further analysis. Vegetables were also collected at the same time. Mature, leaves of spinach and tomato were collected in clean polyethylene bags.

Analysis: soil samples were air dried before analysis and sieved through 2mm sieve. Chemical analysis was conducted for several parameters including: pH(pH meter),EC(conductivity meter) organic carbon %(black and walkley method,Nitrate N (Kjeldahl method),Available phosphorus(Olsen's method), Total Cr, Cd, Pb and Ni(Wet digestion). 0.5 gm each of soil sample was digested (wet acid digestion) with concentrated HNO_3, H2SO$_4$ and $HClO_4$ (5:1:1) at 800^{o}C (Allen et al. 1986) until the solution become transparent. Vegetables were thoroughly washed with tap water and then by distilled water to remove the soil particles and other contamination. Then vegetables were cut into small pieces and kept in oven at 110^o C for 24 hours. The dried vegetables were then grounded and passed through 2mm sieve and then 1 gm of each vegetable sample was digested similar to soil samples. The digested samples of soil and vegetables were filtered through the whatman No.42 filter paper and the filtrates were diluted to 50ml with distilled water. All reagents used were Merck, analytical grade (AR) including standard stock solutions of known concentration of different heavy metals. Heavy metal concentrations of soil and vegetable samples were estimated by atomic absorption spectrometer (GBC 932+).

Statistical analysis

Experimental data obtained was further statistically analyzed using ANOVA with the help of SPSS software version 20.The results are reported in mean with the standard deviation.

RESULTS AND DISCUSSION

Table 1: Chemical Characteristics of soil irrigated with wastewater (pre monsoon) n=36

Sites/ Parameters	pH	EC	OC%	Nitrate N (mg/kg)	Available P (mg/kg)	Available potassium Kg/h	Cr (mg/kg)	Cd (mg/kg)	Ni (mg/kg)	Pb (mg/kg)
Mansarovar										
Mean	8.21*	0.98	1.05**	33.56*	20.06**	242.94**	35.60**	12.37**	25.86**	30.03**
SD(±)	0.12	0.02	0.03	2.47	1.23	15.60	1.67	1.31	1.55	1.44
Pratap nagar										
Mean	8.32*	1.02*	0.98*	35.21*	22.44**	252.60**	36.41**	12.39**	24.30**	30.64**
SD(±)	0.31	0.09	0.06	2.41	0.52	13.74	2.35	1.22	1.77	1.42
Control										
Mean	7.52	0.91	0.66	20.95	12.02	154.16	0.001	ND	0.002	ND
SD(±)	0.11	0.04	0.20	1.38	0.26	27.42	0.01	--	0.01	--
Safe limits[a]	--	--	--	--	--	--	--	3-6	75-150	250-500

[a] Awashthi 2000, * significant at 0.05 level,**significant at 0.01 level, ND- not detected,

The results for soil analysis for pre-monsoon (table 1) and post monsoon period (table 2).The results indicate an increase in the nutrients due to application of wastewater at both the sites as compared to the control site where bore well water is used for irrigation. pH values of soil showed a slight difference during pre and post monsoon. The values for pH were found to be higher during pre monsoon as compared to post monsoon.

The increase of soil pH might be due to addition of various soluble salts found in industrial effluent. An increase in soil pH was also reported by [2][14] while working on soil characteristics affected by long term application of sewage wastewater. Such effect may be attributed to the high content of basic cations *viz.* Na^+, Ca^{2+} and Mg^{2+} in the wastewater, which after accumulation in surface soil layer for a long period of time leading to increase in soil pH. A slight decrease in pH was observed during the present study in samples taken post monsoon. This decrease can be attributed to less or no use of wastewater during monsoon reducing the accumulation of salts in the soil.

Table 2:Chemical Characteristics of soil irrigated with wastewater(post monsoon) n=36

Sites/ Parameters	pH	EC	OC%	Nitrate N (mg/kg)	Available P (mg/kg)	Available potassium Kg/h	Cr (mg/kg)	Cd (mg/kg)	Ni (mg/kg)	Pb (mg/kg)
Mansarovar										
Mean	7.89*	0.95*	0.91*	25.66**	17.35**	188.97**	27.41**	9.98**	23.22**	19.78**
SD	0.02	0.00	0.22	0.01	0.45	0.02	0.01	0.01	0.03	0.59
Pratap nagar										
Mean	7.92*	0.97*	0.87*	27.73**	20.35**	201.80**	30.34**	10.14**	21.51**	21.11**
SD	0.23	0.01	0.01	0.20	0.25	0.21	0.05	0.03	0.02	0.33
Control										
Mean	7.50	0.89	0.67	20.87	11.98	152.66	ND	ND	ND	ND
SD	0.01	0.00	0.01	0.02	0.03	0.05	--	--	--	--
Safe limits [a]	--	--	--	--	--	--	--	3-6	75-150	250-500

[a] Awashthi 2000, * significant at 0.05 level,**significant at 0.01 level, ND- not detected,

Table 3: Mean Concentration of metals in vegetables during pre monsoon period

	Cr (mg/kg)	Cd (mg/kg)	Ni (mg/kg)	Pb (mg/kg)
	Spinach			
Mansarovar	14.21**±1.54	2.28**±0.45	2.96**±0.77	10.22**±0.06
Pratapnagar	16.26**±1.30	4.62**±1.32	5.02**±1.12	12.86**±1.22
	Tomato			
Mansarovar	12.46**±1.54	1.84**±0.68	1.66**±0.87	8.49**±0.24
Pratapnagar	13.78. **±1.20	2.69**±0.59	3.25**±1.47	9.82**±2.44
Control	BDL	BDL	BDL	BDL
Safe limits[b]	0.1-0.2	0.1-0.5	1.50	0.1-0.3

[b] Limits given by WHO/FAO (codex standard 193-1995), ,**significant at 0.01 level

Table 4: Mean Concentration of metals in vegetables during post monsoon period

	Cr (mg/kg)	Cd (mg/kg)	Ni (mg/kg)	Pb (mg/kg)
	Spinach			
Mansarovar	12.98**±0.25	1.21**±0.32	1.89±0.27	7.62**±0.31
Pratapnagar	14.56**±2.01	2.69**±0.45	3.87**±0.55	9.47**±1.24
	Tomato			
Mansarovar	10.02**±0.23	0.97**±0.61	0.64**±0.20	6.31**±0.34
Pratapnagar	11.95. **±1.42	1.28**±0.08	3.25**±1.47	7.89**±1.55
Control	BDL	BDL	BDL	BDL
Safe limits[b]	0.1-0.2	0.1-0.5	1.50	0.1-0.3

[b] Limits given by WHO/FAO (codex standard 193-1995)

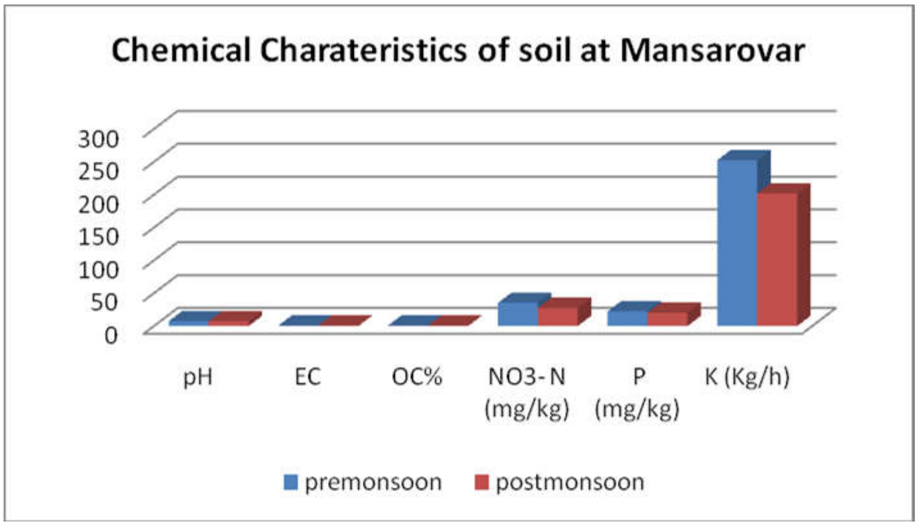

Figure 1:Chemical characteristics during pre and post monsoon period at mansarovar

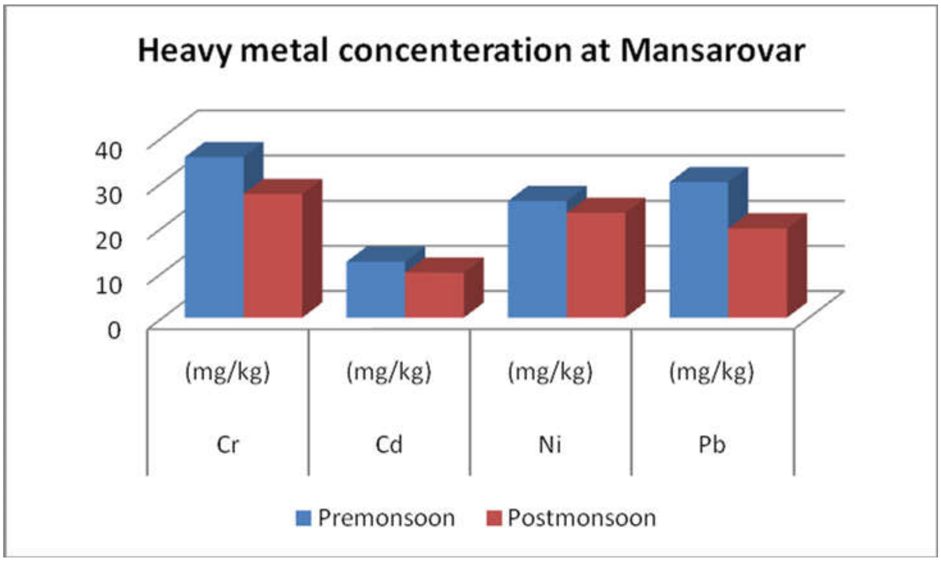

Figure 2:Heavy metal concentration during pre and post monsoon period at Mansarovar

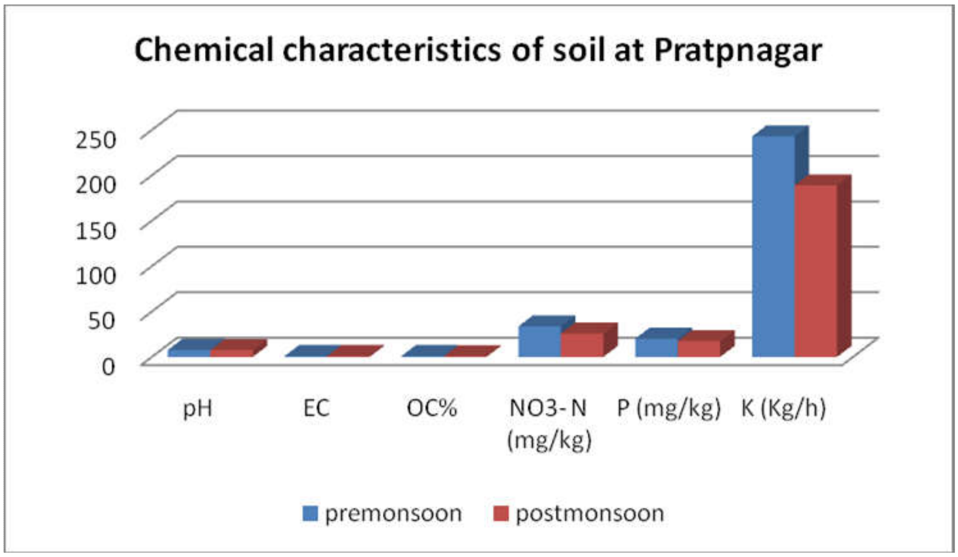

Figure 3:Chemical characteristics during pre and post monsoon period at Pratap nagar

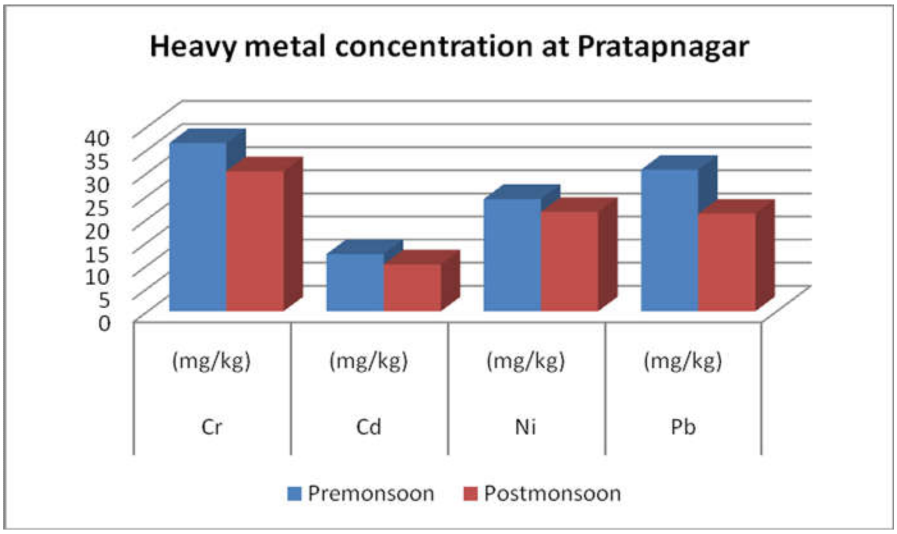

Figure 4: Heavy metal concentration during pre and post monsoon period at Pratapnagar

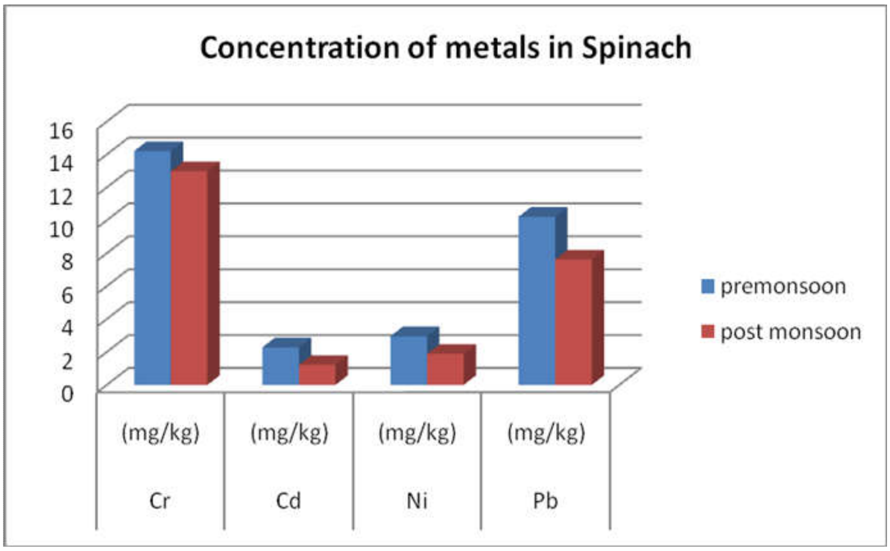

Figure 5: Mean Concentration of heavy metals in Spinach

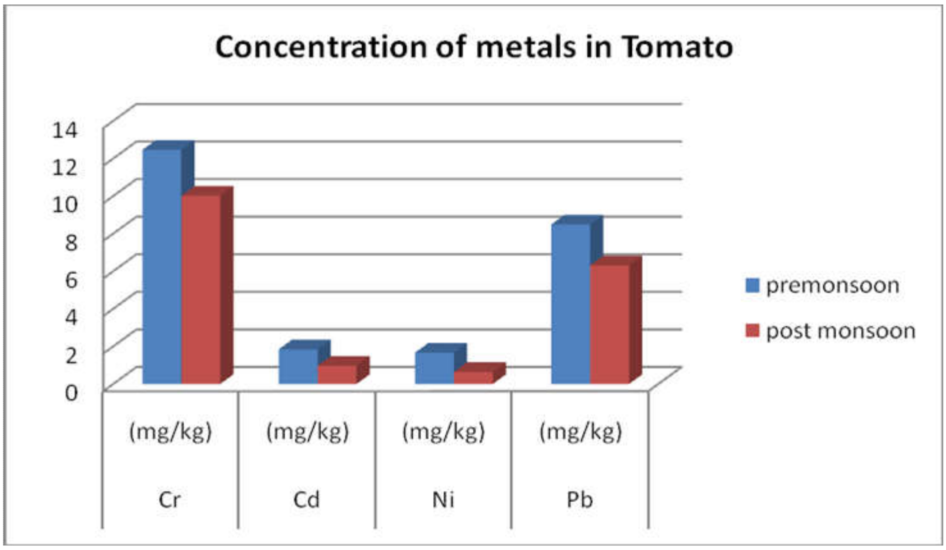

Figure 5:Mean Concentration of heavy metals in Tomato

Electrical Conductivity was also found to increase in the present study with the application of wastewater as compared to bore well irrigated site. The findings of present study are in line with the observations of [9]. The reason for this can be accumulation of dissolved salts found in the industrial wastewater.

Soil organic carbon (OC %) is the most important indicator of soil quality and plays a major role in nutrient cycling and regulating the bioavailability of metals in the soil. Heavy metals are bound to Organic matter making them less bioavailable. In the present study, application of wastewater was found to increase the organic carbon% in the soils as compared to the bore well water irrigated soils during the pre monsoon period. The OC% was found highest (1.05%) in soils of mansarovar as compared to Pratap nagar (0.98%) and control (0.66%).Mansarovar is majorly residential area thus the wastewater from this area contributes more of organic matter in the wastewater. The most important constituents found in sewage are organic matter, nitrogen, phosphorus, carbon, calcium, magnesium, sulfate, chloride, carbonate. Several other research studies have found the similar results as found in the present study. Higher value of OM %(2.85 %) in wastewater irrigated sites as compared to control site (1.98 %) were observed by[5] at Turkey, an Increase of OC % from 22% to 30 % in wastewater irrigated sites was also reported by[11] at Iran. 35.54 % increase in the OC % of wastewater irrigated site as compared to the control site at Dehradun, India was observed by[12]. This increase in OM and OC % can be attributed to the presence of organic matter, nitrogen, phosphorus, carbon, calcium, magnesium, sulfate, chloride, carbonate in the wastewater. In contrast to the results during pre monsoon period, slight decrease in the OC % was found during post monsoon period. The mean OC % at Mansarovar decreased from 1.05 to 0.91% and from 0.98 to 0.87 at Pratap nagar. This can be due to reduction in the addition of wastewater during rainy season.

The mean NO3-N,available P and Potassium concentrations was found to increase in wastewater irrigated site and showed a significant difference at ($p<0.01$) level as compared to the control site during the pre and post monsoon periods. Higher concentration of No3-N,available P and Potassium were found at Pratap nagar site as compared to Mansarovar .Slight decrease in NO3-N, available P and Potassium levels were observed during the post monsoon period

The reason for the increase in nutrients as compared to bore well water irrigated soil can be attributed to the presence of Nitrates and phosphates in the wastewater. Several industries including textile industries are a major source of nitrates as well as detergents from domestic effluents contributes to the nitrate content of wastewater. Several studies support the findings of the present study.It was found that wastewater irrigation increased the nitrate nitrogen in the soil by 37.88% as compared to the soil irrigated by ground water [3].[14]found significantly higher available N in the range of (222.65–283.26 Kg/ha), P (19.13–23.92 Kg/ha) and K (322.27–343.25 Kg/ha) in wastewater irrigated soils as compared to crops irrigation with well water in which N ranged from (205.92–288.50 Kg/ha), P (13.66–23.23 Kg/ha) and K (299.41–345.20 Kg/ha) .This trend indicates that sewage water irrigation provides the essential nutrients to the crops as well as improves fertility levels of soil.

Heavy Metal Accumulation in soil

The results for soil analysis for pre-monsoon (table 1) and post monsoon period (table 2) shows accumulation of heavy metals in the soil due to continuous wastewater irrigation. Significant difference at ($p<0.01$) level is observed at wastewater irrigated sites for the heavy metals (Cr, Cd, Ni and Pb) as compared to the control site. The obtained results also show variation in the concentration of metals and other chemical parameters between pre-monsoon and post-monsoon analysis of soil. Higher concentrations of metals was observed at wastewater irrigated sites during pre-monsoon period as compared to post monsoon period. The possible reason for this could be application of large quantities of wastewater during dry season as compared to the rainy season where minimal or no wastewater is applied to the fields reducing the concentration of metals. At control site Cr and Ni were detected in a very low concentration during pre monsoon period whereas Cd and Pb were not detected in both the periods. The concentration of metals (Cr,Cd, and Pb) was found to be higher at Pratapnagar as compared to that found at Mansarovar . Ni(24.30 mg/kg) was found slightly lower at Pratapnagar as compared to Ni(25.86 mg/kg) at Mansarovar. These levels were found to be lower during post monsoon period with metal concentration .All the metals were below the detectable limit at the control site during the post monsoon period (Table 2).

Several studies support the results obtained in the present study, where long term irrigation with the wastewater containing heavy metals leads to the accumulation of these metals in soil. 143 times more accumulation of Cd in sewage effluent irrigated soils around Calcutta[8] over non-sewage irrigated soils, followed by Pb (18.5 times), Cr (5.6 times), and Ni (2.3 times). Much higher level of metals Cr(246-1980 mg/kg), Cd(0.12-4.22 mg/kg),Ni(32.1-94 mg/kg) and Pb(24-38 mg/kg) in wastewater irrigated sites at Panjab was reported [4] as compared to the concentrations of metals found in the present study during both the pre and post monsoon periods (Table 1-2).[7] also observed accumulation of metals Cr (936 mg/kg),Ni(55.6 mg/kg),Pb(94 mg/kg) in agricultural fields under wastewater irrigation for last 30 years as compared to control site where no wastewater was ever used. These levels of heavy metals are much higher than that found in the present study.

Heavy metal accumulation in vegetables

The vegetable samples at both the study sites showed accumulation of heavy metals above the safe limits. A significant difference at($p<0.01$)level was seen at both the sites and vegetables. The concentration was found to be higher during the pre monsoon season as compared to the post monsoon season. Vegetable samples from pratap nagar showed higher concentration of metals as compared to Durgapura. Spinach, a leafy vegetable showed higher accumulation of metals as compared to tomato. Spinach accumulated highest concentration of Cr followed by Pb,Niand Cd at pratap nagar during pre monsoon period. The values of metals in spinach were found to decrease during the post monsoon period(Table 3)(Fig5)In tomato, during pre monsoon, concentration of Cr was followed by Pb, Cd and Ni which further decreased during post monsoon period. Several other studies have also reported accumulation of metals due to wastewater irrigation. Singh *et al*., 2012 reported a much higher value of Pb(23mg/kg) in spinach whereas as compared to the results of present study. Many fold higher concentration of Pb(39.6 mg/kg) in leafy vegetables and Pb(83..7 mg/kg) in fruit vegetables was reported by [13].The mean concentration of Cd in vegetables in the present study in both the study periods is higher than Cd(0.36 mg/kg)reported by [1] at Ghana and Cd(0.073 mg/kg) reported by [15] . [6] conducted a similar study at Nigeria and reported a reduction in metal concentration in vegetables during the post monsoon period. They reported lower level of Cr(0.6 mg/kg),Ni(2.02 mg/kg) and Pb(2.02 mg/kg) in spinach and Cr(0.36mg/kg),Ni(1.20 mg/kg) and Pb(1.79 mg/kg) in tomato during the dry period as compared to the present result and further lower levels during rainy season. The high concentration of metals in soil and vegetable can be attributed to application of large quantities of wastewater during dry season, adding up to more metals whereas during rainy season minimum or no wastewater is used reducing the levels of metals.In addition to this ,rain water also help to leach down the metals from the root zone. The uptake of metals in the crops also is dependent on various factors, which include temperature, moisture, organic matter, pH and nutrient availability and metal concentration in soil. Amongst all factors organic matter and pH plays an important role, where higher pH and OM helps to bind the metals to soil particles making them less bioavailable.

CONCLUSION

The results obtained from the study show an increase in the nutrient status of the soil but simultaneously accumulation of heavy metals (Cd,Cr,Pb and Ni) is also observed in the wastewater irrigated sites. Crops grown on such soil are known to accumulate large quantities of metals at toxic levels which is observed in the vegetable samples obtained in the study area. Several studies have shown that these metals are a cause of several fatal diseases including the disruption of vital systems of the body. Pb adversely affects the nervous system, Cd adversely affects the kidney and liver, Cr affects the cardiovascular, hepatic, renal and neurological systems whereas Ni causes decrease in sperm count, motility and loss of fertility in males. Thus the use of untreated wastewater in agriculture fields is a major concern for human health. It is thus recommended to treat the wastewater prior to use in soil.

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