

INVESTIGATION OF HEAVY METAL STATUS IN SOIL AND VEGETABLES GROWN IN SEWAGE EFFLUENT WATER IRRIGATED SOILS OF ALIGARH: A 5 YEAR STUDY

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ABSTRACT

The present study was conducted for five years (2009-2014) in the Aligarh district to evaluate the levels of different heavy metals (Zn, Cu, Ni, Cr, Pb and Cd) in soils and in vegetables grown on ground water and sewage effluent irrigated soils. The results showed that continuous application of sewage effluent water for a longer period cause's substantial build-up of heavy metals in soils and in the vegetables grown on such soils. The concentration of heavy metals in vegetables obtained from sewage effluent water irrigated soil was in the order Zn > Cu > Cr > Ni > Pb > Cd. Concentration of heavy metals in the vegetables grown varied from plant to plant which may be due to difference in their uptake capability and translocation to edible portion of the plants. The results of present study also indicate that concentration of Cd, Pb and Ni in the vegetables grown on such soils had exceeded the safe permissible limits for human consumption. Health risk index was found >1 for Cd, Ni and Pb indicating a health risk to the local population associated with Cd, Ni and Pb. The study concludes that in order to reduce the health risk and extent of heavy metal contamination regular monitoring of these metals in soils, vegetables is necessary.

Key Words: *Heavy metals, Sewage effluent, Vegetables, plant uptake, soil Contamination*

1. INTRODUCTION

In recent years, due to constraint in availability of fresh water for irrigation, in India waste water is being used for irrigation of agricultural fields in periurban particularly areas. The growing problem of water scarcity has significant negative influence on economic development, human livelihoods, and environmental quality throughout the world. Farmers are mainly interested in general benefits, like increased agriculture production, low cost water source, effective way of effluent disposal, source of nutrients, organic matter etc, but are not well aware of its harmful effects like heavy metal contamination of soils, crops and quality problems related to health. Waste waters are contaminated with trace elements like lead (Pb), copper (Cu), zinc (Zn), boron (B), cobalt (Co) chromium (Cr), cadmium (Cd), arsenic (As), molybdenum (Mo), manganese (Mn) etc. many of which are non-essential and over time toxic to plants, animals and human beings [1]. Long-term application of treated and untreated waste water resulted in significant build-up of heavy metals in soil and in vegetables and

cereals [2-4]. Vegetables absorb heavy metals from soil, air and water, the accumulation of heavy metals in the edible parts of vegetables represents a direct pathway for their incorporation into the human food chain [5, 6]. As vegetables are an important part of human diet (source of protein, vitamins, iron, calcium) it was thought worthwhile to investigate long term effect (5 years) of sewage effluent irrigation on the concentrations of Pb, Cd, Ni, Cr, Zn and Cu in some vegetables grown in soils of periurban areas of Aligarh, and to assess the associated potential health risk to local inhabitants through consumption of these vegetables. Health risk was ascertained through calculation of different hazard quotients.

II. MATERIALS AND METHODS

2.1 Study area

The present study was carried out for five consecutive years (2009-2014) in the periurban areas of Aligarh city ((27°54'30" N, 78°4'26" E) around sewage discharge system (which also contain effluent of small scale industries). Three areas following different irrigation sources demarcated at the experimental sites. Ground water (GW) from deep bore well (>100 m) is used for irrigation at ground water irrigation site (GWI), for the second site sewage effluent water was used partially for irrigation (PWI) whereas sewage effluent water (SW) was used for irrigation at the third site (SWI). Water samples (GW and SW) used for irrigation were collected in a 100 mL polypropylene bottles. One mL HNO₃ was added to these samples to avoid microbial activities. The soil obtained from experimental site was alluvial. The surface soil (0-30 cm depth) was collected, air dried, crushed and grounded to pass through <70 mesh sieve before use. The physico-chemical properties of soils and water samples were determined as discussed earlier [7] and values are given in Table 1.

Edible parts of major vegetables grown in the area were also collected randomly from experimental sites for 5 consecutive years. The collected vegetables were palak (*Beta vulgaris* L. cv. all green), cabbage (*Brassica oleracea* L. var. capitata), cauliflower (*Brassica oleracea* L. var. botrytis), lady's finger (*Abelmoschus esculentus* L.), brinjal (*Solanum melongena* L.), tomato (*Lycopersicon esculentum* L.), bitter melon (*Momordica charantia* L.) and radish (*Raphanus sativus* L.). The samples were washed with tap water to remove soil particles. After washing the vegetables were oven dried at 80° C to constant weight. The dried samples were ground and sieved through a 2 mm sieve and analyzed. The samples were digested in di acid mixture containing HNO₃: HClO₄ (3:1). The digested mixture was heated over a hot plate till brown fumes ceased. It was then dissolved in 5 mL of 2M HCl and the supernatant was analysed for Zn, Cd, Cu, Ni, Cr and Pb by atomic absorption spectrophotometer (Systronics AI1200). Appropriate quality assurance procedure and precautions were taken to ensure the reliability of results. Samples were carefully handled to avoid contamination. Glassware was properly cleaned and reagents were of analytical grade. Deionized water was used throughout the study. Reagent blanks were also used. For validation of the analytical procedure, analyses of the samples were repeated against internationally certified plant standard reference material (SRM-1570), the results were found within ± 2% of standard values.

2.2 Data analyses

AS the vegetables are the sources of human consumption. Therefore the soil to plant transfer quotient is the main source of human exposure. Bioconcentration factor or plant concentration factor (PCF) is a parameter used to describe the transfer from soil to plant edible parts. Plant concentration factor (PCF) was calculated [8] as:
$$PCF = \frac{\text{concentration of metal in edible part}}{\text{concentration of metal in soil}}$$

2.3 Metal pollution index (MPI)

To examine the overall heavy metal concentrations in all vegetables, metal pollution index (MPI) was calculated as [9]:

$$MPI = (C_{m1} \times C_{m2} \times C_{m3} \times C_{m4} \dots \times C_{mn})$$
 where C_{fn} = concentration of metal n in the sample.

2.4 Health risk index (HRI)

The health risk index was calculated as the ratio of estimated exposure of test vegetables and oral reference dose [10]. Oral reference doses were 4×10^{-2} , 0.3 and 1×10^{-3} mg kg⁻¹ day⁻¹ for Cu, Zn and Cd, respectively [11] and 0.004, 0.02 and 1.5 mg kg⁻¹ day⁻¹ for Pb, Ni and Cr, respectively [12]. Estimated exposure is obtained by dividing daily intake of heavy metals by their safe limits. An index more than 1 is considered as not safe for human health [11]. Daily intake was calculated by the following equation:

$$DIM = \frac{(\text{heavy metal concentrations in vegetables (ug g}^{-1}) \times \text{Daily intake of vegetables})}{\text{average body weight}}$$

Data of average daily vegetable consumption was obtained through a formal survey of 25 families each having > 4 members conducted in the studied area [13]. The average body mass was taken 55.9 kg.

III. RESULTS AND DISCUSSION

3.1 Levels of heavy metals in water samples

The concentration of heavy metals (ug mL⁻¹) in sewage effluent water ranged from 1.04 to 1.50 for Zn, 1.44 to 2.18 for Cu, 0.34 to 0.416 for Ni, 0.42 to 0.58 for Cr, 0.34 to 0.43 for Pb and 0.11 to 0.148 for Cd (Table 1). The concentration of heavy metals in sewage effluent water was higher by 3.8 to 5.7 times than in ground water. It was found that mean value of Cu, Ni, Cr and Cd in the sewage water effluent exceeds the recommended level. The higher concentration of heavy metals in sewage effluent water may be due to discharge of untreated effluent of small scale industries such as electroplating, metal surface treatment and battery. Similar results are also reported by other workers [14, 15].

3.2 Levels of heavy metals in soil samples

The concentration of heavy metals (ug g⁻¹) in soils irrigated partially by sewage effluent ranged from 41.2 to 66.4 for Zn; 16.2 to 21.3 for Cu; 11.8 to 18.2 for Ni; 17.8 to 26.4 for Cr; 13.0 to 19.5 for Pb and 0.312-0.382 for Cd, while in sewage effluent water it ranged from 53.1 to 81.2 for Zn; 20.0 to 27.8 for Cu; 16.6 to 21.8 for Ni; 24.4 to 35.4 for Cr; 20.0 to 29.2 for Pb and 0.344 to 0.452 for Cd (Table 1). Heavy metal content in partial

sewage effluent irrigated soils was higher in tune of 1.58 times for Zn; 1.99 for Cu; 1.38 for Ni; 2.20 for Cr; 1.70 for Pb and 2.34 for Cd than ground water irrigated soils, while in sewage effluent irrigated soils was higher in tune of Zn 1.92 times; Cu 2.35; Ni 1.84; Cr 2.80; Pb 2.55 and Cd 2.73 than ground water irrigated soils.. Amongst the studied heavy metals Zn was maximum and Cd minimum. But even the upper limits of heavy metal concentrations were below the permissible limits of PFA standards.

3.3 Heavy metals in vegetables

The heavy metal concentration varied among the vegetables (Fig. 1) which may be attributed to differential absorption capacity of studied vegetables for different heavy metals. Concentration of all the studied heavy metals in vegetables grown in partial sewage effluent irrigated and sewage effluent irrigated soils were several folds higher in vegetables grown in ground water irrigated soils. Similar results were also found by other researchers [4,16]. The concentration of heavy metals in vegetables grown were in order sewage effluent irrigated soils > partial sewage effluent irrigated soils > ground water irrigated soils.

Table 1: Mean values of heavy metals content in sewage effluent, ground water (mg L⁻¹) and in soils (mg kg⁻¹) irrigated by sewage effluent and ground water.

Metals	Water		Permissible limit (Indian standard)	Soil			
	Ground water	Sewage effluent water		Ground water irrigated	Partial Sewage effluent water irrigated	Sewage effluent water irrigated	Permissible limit (Indian standard)
Zn	0.340±0.034 (0.24-0.41)	1.28±0.06 (1.04-1.50)	2.0	34.0±0.36 (32.1-37.6)	53.8±1.7 (41.2-66.4)	65.3±1.9 (53.1-81.2)	300
Cu	0.438±0.026 (0.40-0.52)	1.86±0.15 (1.44-2.18)	0.20	9.8±0.11 (8.7-11.4)	19.5±0.86 (16.2-21.3)	23.0±0.96 (20.0-27.1)	140
Ni	0.060±0.002 (0.05-0.086)	0.376±0.015 (0.34-0.416)	0.1	10.34±0.068 (9.14-12.54)	14.3±0.32 (11.8-18.2)	19.0±0.26 (16.6-21.8)	75
Cr	0.080±0.003 (0.06-0.097)	0.504±0.027 (0.42-0.58)	0.2	10.16±0.14 (8.94-11.82)	22.3±0.92 (17.8-26.4)	28.4±0.85 (24.4-35.4)	100
Pb	0.084±0.003 (0.07-0.11)	0.388±0.016 (0.34-0.43)	5.0	9.42±0.046 (8.62-10.48)	16.0±0.64 (13.0-19.5)	24.0±0.62 (20.0-29.2)	300
Cd	0.024±0.002	0.128±0.006	0.01	0.146±0.002	0.342±0.006	0.398±0.006	3

	(Tr-0.04)	(0.11-0.148)		(0.105-0.170)	(0.312-0.382)	(0.344-0.452)	
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The mean concentration of Zn was highest in all the studied vegetables (Table 2) (Fig. 1) it ranges from 3.9 to 14.3; 19.4 to 44.6; 29 to 65.2 $\mu\text{g g}^{-1}$ for ground water irrigated, partial sewage water irrigated and sewage water irrigated water respectively. The concentration of Zn was maximum in cabbage and minimum in bitter gourd. The maximum concentration of Cu and Cr in both partial sewage irrigated and sewage irrigated soils was found in lady's finger and minimum in bitter gourd. The maximum concentration of Ni was in palak and minimum in lady's finger; the maximum concentration of Pb was in cauliflower and minimum in cabbage. The Cd was most in cabbage and least in cauliflower. The concentration of metals in vegetables followed the order $\text{Zn} > \text{Cu} > \text{Pb} > \text{Ni} > \text{Cr} > \text{Cd}$ (Table 2). Variation in heavy metal concentrations in edible parts of vegetables may be due to variation in absorption of metals in plants through roots and their further translocation within the plants parts. It was also found that the concentration of Zn in cabbage and lady's finger; Cu in lady's finger; Ni and Pb in all the studied vegetables were higher than the permissible limits of Indian standards. The data of the study also denote a positive significant intra-plant relationship of heavy metals (Table 3). Plant concentration factor (PCF): The PCF for all the metals in ground water irrigated sites was lower (0.068-0.484) than sewage effluent irrigated sites (0.123-1.294). The PCF for Zn, Cu, Cr and Pb was highest for lady's finger. Maximum PCF for Cd was for cabbage, while it was palak for Ni. It was also found that the minimum PCF was for Cr (Table 4). Variations in PCF among different vegetables may be attributed to bioavailability of metals which depends on its soil concentration, chemical forms and differential absorption capacity of vegetables for different heavy metals. Similar results are also reported by Eissen and Douglass [16], Chauhan and Chauhan [17].

Table 2: Concentration of heavy metals ($\mu\text{g g}^{-1}$) in different vegetables grown in ground water, partial sewage effluent and sewage effluent irrigated sites

Metal	Vegetables								
	Palak	Cabbage	Brinjal	Lady's Finger	Tomato	Bitter gourd	Radish	Cauliflower	
Zn	GW1	13.2 (12.2-14.8)	14.3 (13.8-15.3)	10.4 (9.9-11)	11 (10.8-11.4)	12 (11.5-12.4)	3.9 (3.84-4)	15 (14.6-15.5)	8.8 (8.5-9)
	PW1	28.6 (27.1-29.4)	44.6 (43.6-45.5)	21.3 (20.4-22.2)	39.5 (38.9-40.4)	23.1 (22.4-23.6)	19.4 (18.6-20)	29.2 (28.6-30)	26.4 (25.8-26.8)
	SW1	39.5 (38.2-40.4)	65.2 (63.8-66.8)	27.2 (26.4-28.1)	60.5 (57.6-61.4)	34.6 (33.9-35)	29 (27.7-31)	40 (38.9-41)	38.4 (37.9-39)
Cu	GW1	4.3 (3.8-4.8)	2.6 (2.1-3.0)	3.9 (3.5-4.5)	3.6 (3.46-3.76)	4.7 (4.54-5.10)	1.16 (1.07-1.25)	4 (3.7-4.2)	3.4 (3.2-3.9)
	PW1	10.0 (9.2-11.0)	6.5 (5.9-7.0)	9.2 (8.8-9.6)	18.2 (17.2-20.2)	11.5 (11.2-11.8)	5 (4.6-5.5)	10.6 (10.3-10.8)	10.9 (10.3)
	SW1	15.3 (14.5-16.2)	9.4 (8.8-9.9)	16.6 (15.9-17.3)	24.2 (22.8-26.4)	17.2 (16.6-18)	7.8 (7.4-8.3)	14.7 (14.4-15)	13.4 (13.1-13.7)
Ni	GW1	1.05 (0.96-1.14)	0.50 (0.46-0.55)	0.70 (0.66-0.74)	0.37 (0.33-0.40)	0.70 (0.68-0.72)	0.34 (0.32-0.37)	0.65 (0.62-0.67)	0.52 (0.5-0.55)
	PW1	9.1 (8.4-9.6)	6.8 (6.2-7.4)	8.2 (7.8-8.5)	4.4 (4.24-4.56)	8.7 (8.64-8.92)	7.5 (7.34-7.62)	10.4 (10.1-10.8)	9.7 (9.5-10)
	SW1	15.4 (14.6-16.1)	11.1 (10.8-12.0)	15.3 (14.4-16.0)	6.8 (6.62-7.10)	14 (13.5-14.3)	12 (11.2-12.6)	14.4 (13.9-15)	14.0 (13.6-14.5)
Cr	GW1	0.74 (0.66-0.80)	0.45 (0.4-0.49)	0.46 (0.42-0.48)	0.28 (0.25-0.30)	0.74 (0.72-0.76)	0.23 (0.21-0.26)	0.64 (0.60-0.66)	0.73 (0.68-0.77)
	PW1	4.44 (4.14-4.82)	4.78 (4.44-4.90)	5.86 (5.68-6.0)	8.66 (8.34-8.9)	7.73 (7.64-7.88)	2.76 (2.64-2.9)	5.5 (5.44-5.6)	2.95 (2.88-3)
	SW1	8.12 (7.84-8.36)	6.54 (6.24-6.78)	8.3 (7.9-8.6)	12 (11.4-12.8)	13 (10.6-11.3)	4.54 (4.42-4.64)	7.95 (7.82-8.06)	4.6 (4.3-4.9)
Pb	GW1	0.54 (0.48-0.60)	0.40 (0.38-0.42)	0.76 (0.73-0.80)	0.33 (0.31-0.35)	0.57 (0.54-0.60)	0.15 (0.14-0.16)	0.50 (0.5-0.54)	0.75 (0.73-0.78)
	PW1	9.9 (9.4-10.5)	7.0 (6.6-7.5)	10.2 (9.5-10.7)	11.8 (11-12.6)	8.2 (7.8-8.5)	7 (6.8-7.3)	6 (5.8-6.3)	12 (11.6-12.3)
	SW1	16.0 (15.5-16.4)	11.9 (11.3-12.5)	16.1 (15.5-16.4)	18.6 (18-19.4)	14.2 (13.6-14.5)	15 (14.5-15.7)	12.4 (12-12.7)	23 (22.8-23.4)
Cd	GW1	0.046 (0.044-0.048)	0.035 (0.032-0.038)	0.037 (0.034-0.038)	0.062 (0.06-0.065)	0.024 (0.023-0.026)	0.01 (0.00-0.01)	0.029 (0.0285-0.0294)	0.026 (0.0255-0.0262)
	PW1	0.226 (0.218-0.232)	0.302 (0.282-0.310)	0.293 (0.284-0.30)	0.212 (0.204-0.22)	0.17 (0.166-0.174)	0.27 (0.266-0.274)	0.172 (0.168-0.176)	0.138 (0.134-0.144)
	SW1	0.344 (0.338-0.350)	0.515 (0.503-0.528)	0.376 (0.368-0.384)	0.298 (0.289-0.306)	0.21 (0.202-0.22)	0.35 (0.346-0.355)	0.256 (0.249-0.26)	0.21 (0.205-0.216)

GWI= ground water irrigated soil; PWI= partial sewage effluent irrigated soil; SWI= sewage effluent irrigated soil

Table 3: Correlation coefficients among heavy metals in vegetables

Metal	Cu	Ni	Cr	Cd	Pb
Zn	0.764	0.724	0.843	0.862	0.776
Cu		0.912	0.884	0.686	0.884
Ni			0.794	0.714	0.744
Cr				0.914	0.912
Cd					0.804

Table 4: Transfer factor of heavy metals through different vegetables at ground water and sewage effluent irrigated sites

Metal		Vegetables							
		Palak	Cabbage	Brinjal	Lady's Finger	Tomato	Bitter gourd	Radish	Cauliflower
Zn	GWI	0.388	0.420	0.306	0.323	0.353	0.115	0.411	0.259
	PWI	0.531	0.829	0.390	0.734	0.425	0.361	0.543	0.491
	SWI	0.734	0.998	0.416	0.919	0.530	0.404	0.615	0.588
Cu	GWI	0.438	0.265	0.398	0.369	0.484	0.118	0.408	0.347
	PWI	0.512	0.334	0.471	0.948	0.590	0.256	0.543	0.513
	SWI	0.652	0.409	0.722	1.053	0.739	0.339	0.639	0.582
Ni	GWI	0.102	0.048	0.068	0.036	0.068	0.033	0.064	0.051

	PWI	0.636	0.475	0.573	0.314	0.6147	0.524	0.727	0.678
	SWI	0.811	0.584	0.806	0.362	0.737	0.632	0.758	0.734
Cr	GWI	0.072	0.045	0.046	0.028	0.074	0.023	0.064	0.073
	PWI	0.199	0.214	0.263	0.388	0.348	0.123	0.247	0.132
	SWI	0.290	0.232	0.293	0.422	0.388	0.160	0.280	0.162
Pb	GWI	0.057	0.042	0.081	0.035	0.061	0.026	0.055	0.080
	PWI	0.618	0.438	0.637	0.737	0.512	0.437	0.375	0.750
	SWI	0.750	0.496	0.671	0.817	0.592	0.625	0.517	0.958
Cd	GWI	0.315	0.240	0.253	0.425	0.164	0.068	0.199	0.178
	PWI	0.684	0.883	0.856	0.620	0.497	0.789	0.502	0.418
	SWI	0.864	1.294	0.944	0.726	0.528	0.879	0.643	0.528

GWI= ground water irrigated soil; PWI= partial sewage effluent irrigated soil; SWI= sewage effluent irrigated soil

Table 5: Health risk index (HRI) for different heavy metals due to consumption of vegetables grown on sewage effluent irrigated soils.

Vegetable	Zn		Cu		Ni		Cr		Pb		Cd	
	PWI	SWI	PWI	SWI	PWI	SWI	PWI	SWI	PWI	SWI	PWI	SWI
Palak	0.92	1.12	1.24	1.39	1.88	2.56	0.14	0.16	1.12	1.24	2.84	3.42
Cabbage	1.24	1.94	0.80	0.82	1.86	2.12	0.124	0.12	0.84	0.66	3.148	4.16
Brinjal	1.18	1.42	1.48	1.74	3.14	3.85	0.24	0.28	1.88	2.14	4.98	5.96

Lady's Finger	1.84	2.25	1.66	1.86	0.70	0.75	0.20	0.22	1.14	1.38	2.76	3.38
Tomato	1.48	1.84	1.32	1.48	3.12	3.86	0.44	0.52	2.14	2.52	4.04	4.58
Bitter gourd	0.36	0.42	0.42	0.45	0.90	0.98	0.10	0.12	0.90	0.96	1.34	1.71
Radish	1.06	1.08	0.74	0.72	1.68	2.04	0.40	0.46	0.88	1.08	2.08	2.46
Cauliflower	1.50	1.84	0.78	0.86	2.68	3.16	0.14	0.18	2.46	2.78	2.38	2.98

PWI= partial sewage effluent irrigated soil; SWI= sewage effluent irrigated soil

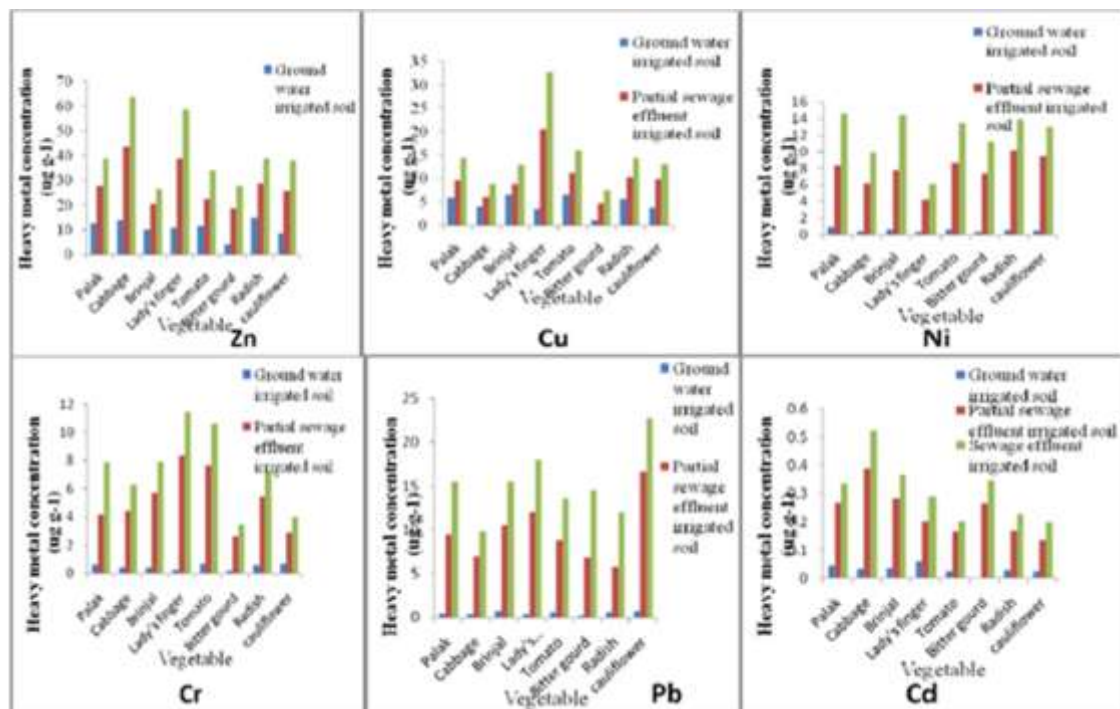


Fig.1. Concentration of heavy metals (ug g⁻¹) in different vegetables

IV. METAL POLLUTION INDEX (MPI)

The values of MPI which helps in metal pollution monitoring of waste water irrigated areas were more than 1 for vegetables grown in both partial sewage irrigated and sewage irrigated soils. The values of MPI for vegetables grown in sewage irrigated soils ranged from 6.34 to 36.9, while ranged from 3.4 to 6.39 for partial

sewage irrigated soils. Among different vegetables brinjal showed highest value of MPI followed by lady's finger. The MPI was least for bitter gourd. The MPI for sewage irrigated vegetables followed the order brinjal>lady's finger>palak>cabbage> tomato>radish> cauliflower> bitter gourd.

Health risk index (HRI): The value of HRI more than 1 indicates a considerable risk of negative impact on human health. Health risk index for Cd was more than 1 for all the studied vegetables (Table 5). For Pb it was higher in palak, brinjal, lady's finger, tomato and cauliflower, whereas for Ni it was higher for all the studied vegetables except lady's finger and bitter gourd. The values of HRI for Cu were more than 1 in palak, brinjal, lady's finger and tomato and for Zn it was more than 1 for all the vegetables except bitter gourd. Health risk index for Cr was less than 1 for all the vegetables. These results showed that Ni, Pb and Cd contamination in plants had potential to pose health risk to consumer.

V. CONCLUSIONS

From the results of present long term study it can be concluded that metal accumulation in the edible part of vegetables mainly depends on their growth habitat like water and soil. Concentration of heavy metals in the vegetables grown varied which may be due to difference in their uptake capability and translocation to edible portion of the plants. Most edible parts of the vegetables grown in sewage water irrigated soils of Aligarh are heavily polluted with heavy metals. Long term consumption of these highly metal contaminated vegetables may cause different disease like brain and kidney damage, cancer, dermatitis etc. Cadmium, Pb and Ni concentration were above the national and international permissible limit in most of the studied vegetables. The data generated may be used as baseline wastewater quality framework to prevent excessive build-up of these heavy metals in food chain. A regular monitoring of heavy metal contamination in the vegetables grown at sewage effluent water irrigated area is necessary and consumption of contaminated vegetables must be avoided.

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