Octa Journal of Environmental Research
International Peer-Reviewed Journal
Oct. Jour. Env. Res. Vol. 4(1): 033-040
Available online http://www.sciencebeingjournal.com

Jan – Mar., 2016
ISSN 2321 3655

Research Article

Octa Journal of Environmental Research
(Oct. Jour. Env. Res.) ISSN: 2321-3655
Journal Homepage: http://www.sciencebeingjournal.com

ASSESSMENT OF VEGETABLES AND SOILS FOR SOME HEAVY METALS FROM IRRIGATED FARMLANDS IRRIGATED WITH INDUSTRIAL EFFLUENTS OF HPC, NAGAON, ASSAM (INDIA)

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Received: 3rd March 2016 **Revised**: 17th March 2016 **Accepted**: 22nd March 2016

Abstract: Metal accumulation in the vegetables/crops growing on agricultural soils, contaminated due to long term irrigation with treated industrial wastewater has been assessed in an area of industrial complex, HPC, Nagaon, Assam (India). To predict the uptake of Cd, Cr, Mn, Ni and Pb by vegetables grown on agricultural soil, samples of 14 varieties of vegetables were collected from seven villages around the study area and the samples were analyzed by Atomic Absorption Spectrophotometer (AAS). Various physico-chemical properties such as pH, electrical conductivity, bulk density, organic carbon, water holding capacity (WHC) and HC of the soil were studied for both pre and post monsoon seasons and the data in the two seasons were compared. The comparison of physico-chemical properties of soil showed that pH, electrical conductivity, and organic carbon, sodium were found high in the pre monsoon season and bulk density, water holding capacity and HC were found high in the post monsoon season. The analysis of metal content in the soil and vegetables showed the order of Mn > Pb > Cr > Ni > Cd and Mn > Pb > Ni > Cr > Cd respectively. The metals were examined for dependency upon some soil factor through the use of correlation analysis, pH, electrical conductivity, organic carbon and HC correlated positively with Cd, Cr, Mn, Ni and Pb indicating that these factors greatly control the concentration of these metals in the soils. Metal transfer factors from soil to vegetable were found less significant for Mn, Ni and Pb, but significant values were obtained for Cd in some sites. The results from the study indicates that there is a need to develop appropriate agricultural management practices to ensure adequate measure of amendment with trace metal concentrations within the critical values for plant growth.
Keywords: Heavy metals; Physico-chemical parameters; Vegetables; Soil.
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INTRODUCTION

Vegetables constitute an important part of the human diet, because they contain important carbohydrates, proteins, vitamins, minerals and trace elements (Dastane, 1987). During the last few years, due to increased awareness on the food value of vegetables, their consumption was increased gradually (Fisseha, 2002). Soil is a very important resource for providing quality food and a livable environment as basic human needs (Wild, 1995). It serves as a sink and recycling factory for both liquid and solid wastes. During the last few decades, as a consequence of industrialization, the heavy metal concentration of soils has increased worldwide (Adriano, 2001). Municipal solid wastes have been found to contain appreciable quantity of heavy metals such as Cd, Zn, Pb, and Cu, all of which may eventually end-up in the soil and are leached down the profile (Alloway and Aryes, 1997). Dumping of municipal or industrial solid wastes is one of the principal sources of heavy metals in the environment. Due to the cumulative behaviour and toxicity, heavy metals in soils have a potential hazardous effect not only on crop plants but also on human health (Das *et al*., 1997). Heavy metals, such as cadmium, lead, and chromium are important environmental pollutants, particularly in areas under irrigated with waste water. Long-term use of waste water for cultivation of leafy and other vegetables has resulted in the accumulation of heavy metals in the soil and their transfer to the various crops under cultivation, with levels of contamination that exceed permissible limits (Nrgholi, 2007). In this respect, contamination of agricultural soils with heavy metals has always been considered as a critical challenge to the scientific community (Faruk *et al*., 2006). Trace elements along with other pollutants are discharged into the environment through various activities which are taken up by animals and plants from the environment through air, water and food. Thus, there is the need to carry out extensive assessment on heavy metal sources, their accumulation in the soil and the effect of their presence in water and soil on plant systems.

In the present investigation, an attempt has been made to study the distribution pattern of metals (Cd, Cr, Mn, Ni and Pb) in soil and in fourteen different vegetables, namely Brinjal, V1, (*Solanum melongena* Linn.), Cabbage, V2, (*Brassica oleracea* Linn. Var. *capitata* Linn.*),* Cauliflower, V3, (*Brassica oleracea* Linn. Var. *botrytis* Linn.) French-bean, V4, (*Phaseolus vulgaris* L.), Knolkhol, V5, (*Brassica oleracea* Linn. Var. *gongylodes* Linn.), Papaya, V6, (*Carica papaya* Linn.), Potato, V7, (*Solanum tuberosum* Linn.*),* Radish, V8, (*Raphanus sativus* Linn.), Coriander, V9, (*Amaranthus spinosus),* Mosondari, V10, (B. *oleraceae* L.), Spinach, V11, (*Spinacia oleracea* Linn.), Sping speng amaranth, V12, (*Honyttinia cordata)* Tampala, V13, (*Raphanus sativus* L.) and Suka Saak, V14, (*Rumex vesicarius* L.) grown in irrigated soil around industrial area of Jagiroad.

Study area: The study area taken up in this work is a part of the district of Morigaon in Assam (India). The area, known as Jagiroad, surrounds the Nagaon Pulp and Paper Mill–a unit of the Hindusthan Paper Corporation Limited, a Government of India undertaking. Jagiroad is a small township along the national highway, NH 37, almost in the midway between Guwahati in the west and Nagaon in the east at a distance of 50 Km and 70 Km from Guwahati and Nagaon respectively. The study area lies between $26^{0}5^{/}$N and $26^{0}15^{/}$N latitudes and $92^{0}10^{/}$ E and $92^{0}15^{/}$ E longitudes. The fluvial process under the tropical humid climatic regime has given rise to a plain topography in the area with occasional marshes and swamps towards its north, forming the huge wetland in the form of the Taranga Beel. The Sonaikuchi Reserve Forest lies in its southern part, which borders the hills of the Meghalaya plateau.

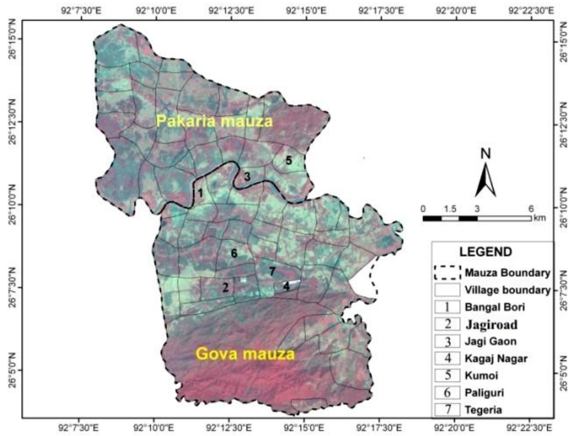

Figure 1. Map showing the Location of Jagiroad (Gova mouza)

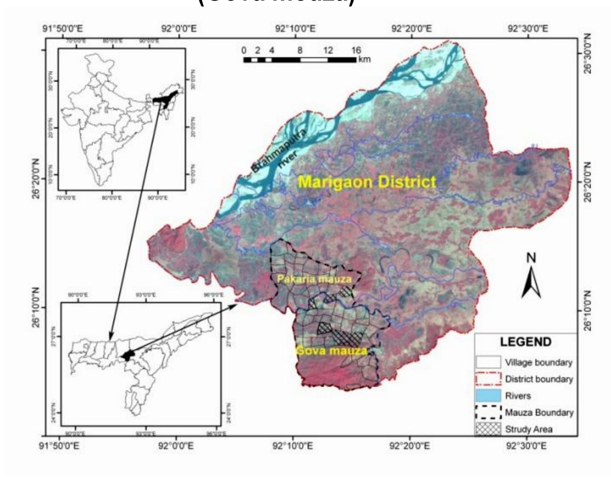

Figure 2. The study area and the seven sampling sites

Drainage: The general slope of the land surface is towards the Northwest. The three rivers, Kolong, Kapili and Killing, which finally drain to the parent river Brahmaputra, control the drainage system of the area at large. These tributaries in the north of the study area, with several rivulets and drainage channels discharging into them carry with them heavy quantities of suspended matter and sediments.

Nagaon Paper Mill and Impact area: Nagaon paper mill, a public sector pulp and paper mill, under the Hindustan Paper Corporation Limited, was commissioned in 1985. Thirteen villages with 5644 acres of cultivable land area have been identified as falling under the pollution impact of Nagaon Paper Mill. These villages are Tegheria, Ghunusa, Nakholagrant , Kuiadal, Gauri pathar, Aujuri(I), Aujuri(II), Bengfar, Dayang, Paliguri, Chakumaku, Bongal Bari and Donga Bari.

EXPERIMENTAL

Samples of soil and vegetables were collected from seven selected sites– four sites (Paliguri, N1; Paliguri, N2; Kagajnagar, E1 and Jagiroad, W1) within the impact zone of Nagaon paper and pulp industry and three more sites (Borbhati, N3; Kumoi, N4 and Bongalbori, NW1) outside this zone. Soil samples were collected by using an auger. In each case, a triangular block was cut with the help of the auger. Six samples were collected from a single station at small distances from one another and these were then mixed together to obtain a composite, representative sample. The vertical soil depth taken for sampling was 5–10 cm from the root zone of the plants. Soil samples were brought to the laboratory in sterile containers and were spread out thinly on a piece of stout paper for drying in air in a shade. The soil samples were covered with a superfine wire net to prevent the entry of dust particles from air. The big lumps were broken down, and plant roots, pebbles and other undesirable matter were removed. Dried soil samples were sieved to obtain 300 mesh fractions**.** The samples were preserved in clean polythene bags for analysis. The soil pH was measured by using digital pH meter (Elico L1 120) in 1:5 soil/water suspensions using standard buffers for calibration. The electrical conductivity (EC) of the soil samples was determined by using a conductivity bridge (Elico CM180) with a conductivity cell (Cell constant 1.0) in 1:5 soil/water suspensions. The WHC was determined by Circular brass box method and HC was determined by using Darcy's law. The Organic Carbon (OC) of the soil samples was determined by tritrimetric method (Walkey and Black, 1974). To determine metal content a solution was prepared by digesting 1 g of soil with a triple acid mixture (1: 2: 4) HCl-HNO_3-H_2SO_4. The residue was treated with 6N HNO_3. It was then filtered with Whatman 42 filter paper and made up to 50 ml. The concentration of the metals (Cd, Cr, Mn, Ni, and Pb) in that solution was measured with AAS (Perkin Elmer Analyst200). The vegetable samples were collected randomly by hand, carefully packed into polythene bags and brought to the laboratory for further analysis. These vegetables were carefully washed with tap water to eliminate the adhering soil and other contaminants. They were then dried in an air-oven at ~80^{o}C, and then crushed and mixed thoroughly for the processing of metal analysis. All the samples were analyzed in three replicates. Known amounts of the dried samples were calcined in a muffle furnace at ~600^{o}C to prepare the ash. The ash was taken for analysis of Cd, Cr, Mn, Ni, and Pb. 1.0 g of calcined ash of each sample was digested with triple acid mixture (1:2:4) HCl.-HNO_3-H_2SO_4. The residue was treated with 6N HNO_3. It was then filtered with Whatman 42 filter paper and made up to 50 ml. Analysis of Cd, Cr, Mn, Ni, and Pb was carried out with AAS (Perkin Elmer Analyst 200).

RESULTS AND DISCUSSION

Soil physicochemical properties: The pH is an important parameter for characterizing soil and it provides the most rational basis for managing soil for selective agricultural crops, pasture cultivation, forestry, etc. This parameter also provides us information about the potency of toxic substances present in soil. The data on various physico-chemical parameters (pH, EC, Bulk density, Organic Carbon, WHC and HC) of soil collected from seven sites irrigated with treated effluent of the mill along with control are presented in Table 1. The analysis of the data revealed that the mean values of pH of soil samples decreases in the order N2>N1>E1>W1>N3>NW1>N4 in both pre and post monsoon seasons. Soil of the three sites N2, N1 and E1 was alkaline (pH>7) in nature. These sites are within 2 Km^2 area of the mill and the Taranga beel near these sites carries alkaline effluents released from the mill. Soil of the other four sites was all acidic in nature (pH<7). The electrical conductivity (EC) of soil is a measure of the salinity of the soil.

Table 1. Physico-chemical parameters

Site	Pre-monsoon						Post-monsoon				
	pH	EC (ms/cm)	BD (g/cm)	OC (%)	WHC	HC (cm/min)	pH	EC (ms/cm)	BD (g/cm)	OC (%)	WHC
N1	7.85	0.675	0.97	2.07	44.16	0.598	7.65	0.670	1.10	1.96	45.77
N2	8.00	0.300	0.97	1.97	36.09	0.515	7.75	0.310	1.11	1.67	43.32
N3	6.45	0.235	1.29	1.22	49.24	0.419	6.25	0.210	1.45	1.15	50.97
N4	5.85	0.128	1.34	1.19	52.76	0.402	5.70	0.112	1.39	1.09	53.93
NW1	6.06	0.110	1.27	1.20	52.26	0.346	5.80	0.096	1.29	1.16	55.30
E1	7.45	0.295	1.02	1.89	49.77	0.423	7.10	0.288	1.13	1.76	50.16
W1	6.95	0.325	1.19	1.69	45.81	0.422	6.60	0.310	1.20	1.66	52.61

(BD-bulk density, OC- organic carbon)

The analysis of the data revealed that the mean values of EC (Table 1) were found significantly high at the sites (highest in N1 followed by N2 and W1 respectively) receiving wastewater of the mill compared to control. There was no difference in the level of Bulk density, Organic Carbon, WHC and HC amongst various sites (Table 1).

Concentration of heavy metals in soil and vegetable samples: The total metal (Cd, Cr, Mn, Ni and Pb) contents were estimated in all the samples collected from different sites (Table 2). The level of Cd ranged from 0.30-1.20 mg/kg and 0.35-1.13 mg/kg Dry Weights (DW) for soil and vegetable samples respectively. The similar ranges for the metals Cr, Mn, Ni and Pb are 3.20-20.85 mg/kg and 0.65-1.57 mg/kg, 30.00–225.20 mg/kg and 3.40-7.45 mg/kg, 2.60–8.85 mg/kg and 0.64-3.01 mg/kg and 15.00-40.25 mg/kg and 0.62-3.60 mg/kg respectively. The analyzed data showed that metal content in all the soil samples were below the critical values of for plant growth (Cd, 8.0 mg/kg; Cr, 75.0 mg/kg; Ni, 100 mg/kg; Pb, 200 mg/kg) (Linzon, 1978). From a similar investigation, Bvenura *et.al* (2012) reported that the concentrations of heavy metals in the vegetables were in the range of 0.01 mg/kg –1.12 mg/kg dry weight for Cd, 0.92 mg/kg –9.29 mg/kg for Cu, 0.04 mg/kg – 373.38 mg/kg for Mn and 4.27 mg/kg – 89.88 mg/kg for Zn. The results from the analysis of soils revealed that Cd in soil was in the range of 0.01 mg/kg–0.08 mg/kg, Cu levels were 4.95 mg/kg–7.66 mg/kg, Pb levels were 5.15 mg/kg–14.01 mg/kg and Zn levels were 15.58 mg/kg – 53.01 mg/kg. Farooq *et al.,* (2008), Singh *et al.,* (2010), Yadav *et al.,* (2013) and Geetanjali, (2014) had reported that high concentration of Cd in vegetables might be a threat for the consumers.

The concentrations of the metals Cd, Cr, Mn, Ni and Pb in soil and vegetable samples were N1>N2 >W1>E1>NW1>N3>N4 and NW1 >N1>N2>E1>W1>N4>N3 (Figure 3), E1> N2>N1>W1=N4>N3>NW1 and W1>E1 >N3>N4>NW1>N1>N2 (Figure 4), N1 >W1>E1> N2 >N3>N4 >NW1 and E1>W1>N2 >N3>NW1>N4>N1 (Figure 5), N1>W1>N2> NW1>E1>N3>N4 and N2>N1>E1>N3> NW1>N4>W1 (Figure 6) and E1>N1>W1> N2>N3>N4>NW1 and N1>W1>E1 >NW1>N4>N2>N3 (Figure 7) respectively. The average levels of Cd, Cr, Mn, Ni and Pb were found more in the soil samples collected from effluent irrigated sites which may be due to the presence of metals in treated effluent mixed before irrigation than in control soil. The trend in occurrence of heavy metals in soil under study and control areas revealed that Mn>Pb>Cr>Ni>Cd. Out of all the metals studied, the maximum accumulation of Mn and minimum accumulation of Cd was recorded in all the vegetables following the trend of Mn >Pb > Ni > Cr > Cd. The relationship was also examined for dependency upon some soil factor through the use of correlation analysis (Table 3).

Table 2. Metal content in soil and vegetable samples (Mean values in mg/kg)

Site	Soil					Vegetable				
	Cd	Cr	Mn	Ni	Pb	Cd	Cr	Mn	Ni	Pb
N1	1.20	8.20	225.20	8.85	35.15	0.61	0.84	3.83	2.04	3.60
N2	0.95	10.35	100.00	5.50	25.00	0.58	0.65	4.42	3.01	1.09
N3	0.32	4.00	95.00	2.65	20.10	0.35	1.31	4.11	1.30	0.62
N4	0.30	5.00	90.00	2.60	18.00	0.41	1.18	3.40	0.70	1.11
NW1	0.55	3.20	30.00	3.25	15.00	1.13	1.13	4.00	1.13	1.13
E1	0.80	20.85	105.00	2.85	40.25	0.48	1.41	7.45	1.44	1.19
W1	0.90	5.00	170.00	8.25	30.68	0.45	1.57	5.19	0.64	2.35
Min	0.30	3.20	30.00	2.60	15.00	0.35	0.65	3.40	0.64	0.62
Max	1.20	20.85	225.20	8.85	40.25	1.13	1.57	7.45	3.01	3.60
Mean	0.72	8.09	116.46	4.85	26.31	0.57	1.16	4.63	1.47	1.58
STDEV	0.34	6.16	62.89	2.72	9.39	0.26	0.32	1.36	0.83	1.03
Percentile	1.13	17.70	208.64	8.67	38.72	0.97	1.52	6.77	2.72	3.23

Table 3. Correlation between metal content (Pre and post monsoon) and Physico-chemical parameters of soil

Correlation	Pre monsoon					Post monsoon				
	Cd	Cr	Mn	Ni	Pb	Cd	Cr	Mn	Ni	Pb
pH	0.88	0.59	0.59	0.59	0.74	0.86	0.56	0.60	0.59	0.71
EC (ms/cm)	0.83	0.23	0.91	0.79	0.69	0.85	0.26	0.90	0.80	0.70
BD (g/cm)	-0.88	-0.67	-0.52	-0.52	-0.75	-0.94	-0.61	-0.63	-0.63	-0.74
OC%	0.93	0.63	0.68	0.66	0.83	0.95	0.59	0.76	0.73	0.89
WHC	-0.69	-0.18	-0.40	-0.57	-0.30	-0.68	-0.40	-0.49	-0.45	-0.46
HC (cm/min)	0.76	0.22	0.79	0.69	0.53	0.77	0.28	0.80	0.68	0.58

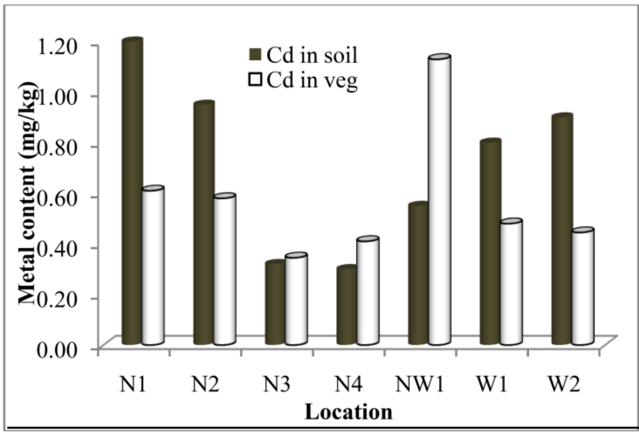

Figure 3. Cd in Soil and Vegetable samples

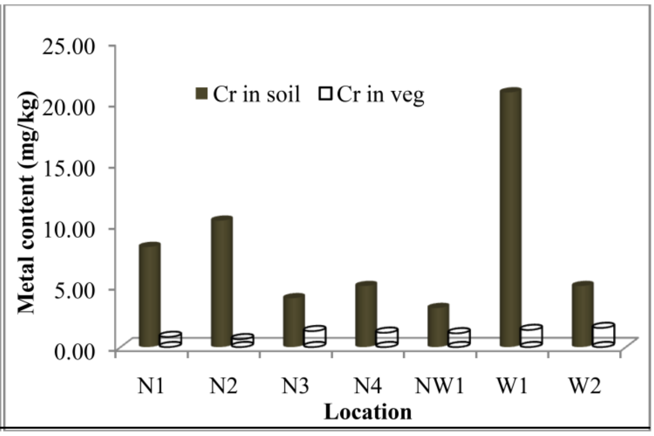

Figure 4. Cr in Soil and Vegetable samples

The pH, Electrical Conductivity, Organic Carbon and HC correlated positively with Cd, Cr, Mn, Ni and Pb indicating that these factors largely control the concentration of these metals in the soils. Negative correlation was found between concentrations of all heavy metals in soil with EC and WHC.

Relationships between the heavy metals in soil and vegetable samples: Correlation coefficients between the concentration of heavy metals in soil and vegetables are presented in Table 4. The Cd concentration in soil correlated positively with the contents of Cd, Mn, Ni and Pb in vegetables. Also, positive correlation were observed between Cr in soil with Cr, Mn, and Ni in vegetables, Mn in soil with Mn, Ni and Pb in vegetables, Ni in soil with Cr, Ni and Pb in vegetables and Pb in soil with Cr, Mn, Ni and Pb in vegetables. Negative correlations were found between Cd in soil with Cr in vegetables, Cr in soil with Cd and Pb in vegetables, Mn in soil with Cd and Cr in vegetables, Ni in soil with Cd and Mn in vegetables and Pb in soil with Cd in vegetables. The positive relationships among the soil and vegetable content metals might be a cause of heavy metals toxicities to plant and animals through their entry into food chain

(Shriadah, 1999). Negative correlation observed among the soil and plant metals content might be due to soil types, nature of plant, extent and type of industrial effluents etc.

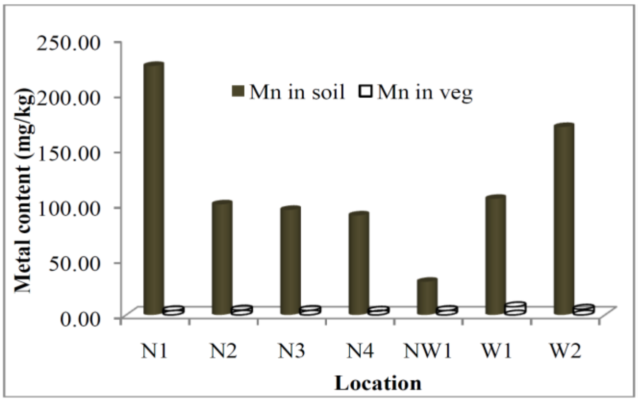

Figure 5. Mn in Soil and Vegetable samples

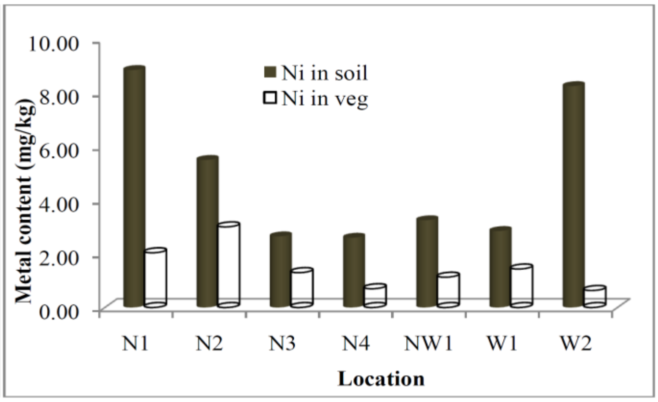

Figure 6. Ni in Soil and Vegetable samples

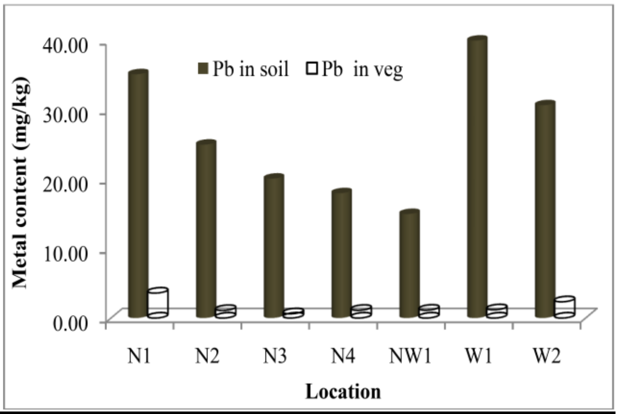

Figure 7. Pb in Soil and Vegetable samples

Transfer factors for heavy metals: Table 5 shows the Transfer Factor (TF) of Cd, Cr, Mn, Ni and Pb from soil to plant. TF is one of the key components of human exposure to metals through the food chain. Transfer factors were determined for Cd, Cr, Mn, Ni and Pb to quantify the relative difference in bioavailability of metals to plants or to identify efficiency of plant species to accumulate a given metal. Absorption and accumulation of heavy metals in plant tissue depend upon factors such as moisture, organic matter, pH and nutrient availability (Sharma *et al,* 2007). However, the physiological response of each plant do exerts control on the degree of bioaccumulation of different metals.

Table 4: Correlation between metal content in soil and in vegetable samples

Correlation	Veg Cd	Veg Cr	Veg Mn	Veg Ni	Veg Pb
soil Cd	0.101	-0.371	0.262	0.548	0.773
soil Cr	-0.212	0.012	0.848	0.340	-0.016
soil Mn	-0.411	-0.072	0.024	0.147	0.885
soil Ni	-0.028	0.234	-0.096	0.234	0.891
soil Pb	-0.342	0.126	0.724	0.218	0.539

Table 5: Transfer factors

Transfer factor	Cd	Cr	Mn	Ni	Pb
N1	0.51	0.10	0.02	0.23	0.10
N2	0.61	0.06	0.04	0.55	0.04
N3	1.08	0.33	0.04	0.49	0.03
N4	1.37	0.24	0.04	0.27	0.06
NW1	2.05	0.35	0.13	0.35	0.08
E1	0.60	0.07	0.07	0.50	0.03
W1	0.49	0.31	0.03	0.08	0.08

The transfer factors as suggested by Kloke *et al*., (1984) for Cd and Zn (1-10) were used as a generalized range for comparisons. These factors were based on the root uptake of metals and discount the foliar absorption of atmospheric metal deposits. In this study all the metals exhibit bioaccumulation in the vegetables with TF< 1 i.e. below Cd and Zn factor range as suggested by Kloke *et al*. (1984) except that for Cd in three sites viz. N3, N4 and NW1 (Table 5) with TF values of 1.08, 1.37 1nd 2.05 respectively. The reasons could be due to the nature of Cd, since Cd is known to be less retained by the soil (Figure 3) than other toxin cations (Chandrappa and Lakeshwari, 2006) or the low organic matter contents of soils sampled could have also enhanced soil-plant transfer of metals.

CONCLUSION

The present study provides the information regarding metal content in soil irrigated with fresh water and treated effluent of HPCL, Nagaon, Assam, India. Metal accumulation in the edible parts of the vegetables growing in those soils was also studied. The study concluded that irrigation of vegetables using treated effluent of HPCL, Nagaon had increased contamination of Cd and Ni in vegetables causing potential health risk in the long term. The average levels of Cd, Cr, Mn, Ni and Pb were found more in the soil samples collected from effluent irrigated sites than in control sites. Out of all the five studied metals, the maximum accumulation of Mn and minimum accumulation of Cd was recorded in all the vegetables. Such variation in the degree of accumulation can be attributed to factors such as plants physiological response, soil characteristics, metal mobility and bioavailability etc. There is thus a need to evolve appropriate agricultural management practices that will ensure adequate measure of amendment with trace metal concentrations within the critical values for plant growth.

Acknowledgment: The authors are thankful to the Department of Chemistry, Gauhati University, Guwahati, Assam for providing necessary laboratory facilities.

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Source of financial support: None.
Conflict of interest: None. Declared.