Investigation of Heavy metal status in soil and vegetables grown in urban area of Allahabad, Uttar Pradesh, India

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Abstract- The present study was carried out to assess levels of different heavy metals like Iron, Cadmium, Nickel, Lead, Copper and Zinc in vegetables irrigated with water from different sources industrial area of Naini Allahabad. The order of heavy metal concentration was found in Fe > Zn > Cd > Pb > Ni > Cu in irrigated water and Fe > Ni > Zn > Cu > Cd > Pb was observed in industrial contaminated sites of soils. Metal levels observed in these sources were compared with WHO. The concentrations (mg/L) of heavy metals in irrigated water ranged from 0.249 to 0.257 for Fe, 0.049 to 0.056 for Zn, 0.028 to 0.036 for Cd, 0.015 to 0.019 for Cu, 0.035 to 0.042 for Pb and 0.031 to 0.038 for Ni which is lower than recommended maximum tolerable levels proposed by joint FAO/WHO Expert committee on food additives (2007), with the exception of Cd and Fe which exhibited elevated content. Uptake and translocation factor of heavy metal from soil to Edible parts of vegetables were quite distinguished for almost all elements examined. Although the practice of growing leafy vegetables using wastewater for irrigation is aimed at producing socio-economic benefits but study reveals that heavy metal-contaminated vegetables grown in wastewater-irrigated areas may pose Public health hazards which is not safe and may not be sustainable in the long-term.

Index Terms- Heavy metal, Wastewater irrigation, Contamination, Bioconcentration Factor.

I. Introduction

Use of wastewater to irrigate agricultural lands is one of common practice in suburban and industrial areas in many parts of the world (Sharma et al. 2007, Gupta et al. 2008). Waste water irrigation leads to accumulation of heavy metals in the soil (Singh et al. 2004, Mapanda et al. 2005, Sharma, Agarwal and Marshall 2007). Sewage waste has been implicated as a potential source of heavy metals such as Copper (Cu), Cadmium (Cd), Zinc (Zn), Lead (Pb), Nickel (Ni) and Iron (Fe) in the edible and non-edible parts of vegetables (Sharma, Agarwal and Marshall 2006). Food safety issues and potential health risks make this as one of the most serious environmental concerns. There is evidence to indicate that agricultural soil also have increased levels of heavy metals as a results of increased in anthropogenic activities (Mc Laughlin and Singh 1999, Sharma Agrawal, and Marshall 2007). Wastewater carries appreciable amounts of trace toxic metals (Pescod 1992, Yadav et al. 2002) which often leads to degradation of soil health and contamination of food chain mainly through the vegetable grown on such soils (Rattan, et al. 2002). The toxic elements accumulated in organic matter in soils are taken up by growing plants and lastly exposing humans to this contamination (Khan et al. 2008).

Municipal solid waste application in agricultural land, wastewater discharge from industries and houses along with abundant pesticide use are the other major sources of the toxic heavy metals. Heavy metals contaminants can be found on the surface and in the tissues of fresh vegetables (Arif et al. 2011). Certain trace elements are essential in plant nutrition, but plants growing in a polluted environment can accumulate trace elements at high contaminations causing a serious risk to human health when they are consumed (Voutsa et al. 1996). Tricopoulos in 1997 revealed carcinogenic effects of several heavy metals such as Cadmium (Cd), Iron (Fe), Lead (Pb), Mercury (Hg), Zinc (Zn) and Nickel (Ni). There have been a number of studies which reported the deposition of heavy metals in soil, Crops and vegetables grown in the vicinity of industrial areas (Yang et al. 2004, Grytsyuk et al. 2006, Mingorance et al. 2007 and Khan et al. 2008).

II. Research Methodology

The Study

The current study was carried out from December 2012 to June 2013 in the around Allahabad industrial processing zone (AIPZ), Naini (25°23'46"N 81°52'17"E) is a suburban industrial town located in Allahabad district, Uttar Pradesh, India. Industrial area comprised of number of industries such as Alstom, ITI Limited, Bharat Pumps & Compressors (headquarters), Areva, Steel Authority of India Limited (SAIL), Dey's medical, Food Corporation of India (FCI), Triveni Structural's Limited (TSL), Cotton Mills subsidiary of Central Government, Raymond polyester plant Most of the people located in this area are industrial workers and belong to the weaker section of the society.

Our study area is situated in the south side of the river Ganga 9 km south of Allahabad city. Naini Allahabad municipality has a conventional sewage treatment plant and final treated effluent from the treatment plant is directly taken to the adjoining agricultural land. The common vegetable grown in the study area is tomato, spinach, coriander, radish, cabbage etc. which are supplied to the all vegetables market in Allahabad and the rest entire the common market. In this study we investigated the concentration of Pb, Zn, Cd, Fe, Cu and Ni irrigation wastewater, soil, and vegetable grown in the mawaiya drain agricultural land area having long term uses of the treated and untreated wastewater for irrigation. The levels of contamination were compared with the Indian Standard guidelines to assess the potential hazards of heavy metals to public health. Most of the industries discharge their effluents without any prior treatment, through open drain which contaminates water, soil and vegetables of the adjoining areas. Different kinds of vegetables such as Tomato (*Lycopersicon esculentum*), Spinach (*Spinacia oleracea*), Coriandrum (*Coriandrum sativum*), Cabbage (*Brassica oleracea*), and Radish (*Raphanus sativum*) are grown in the area throughout the year and are used for home consumption and mainly for selling to residential areas of Allahabad. Samples of soils and these vegetables were collected randomly in triplicate from effluent-contaminated agricultural land located beside Mawaiya drain (a drain in which all the complex industrial effluents from the Allahabad export processing zone are disposed (AEPZ).

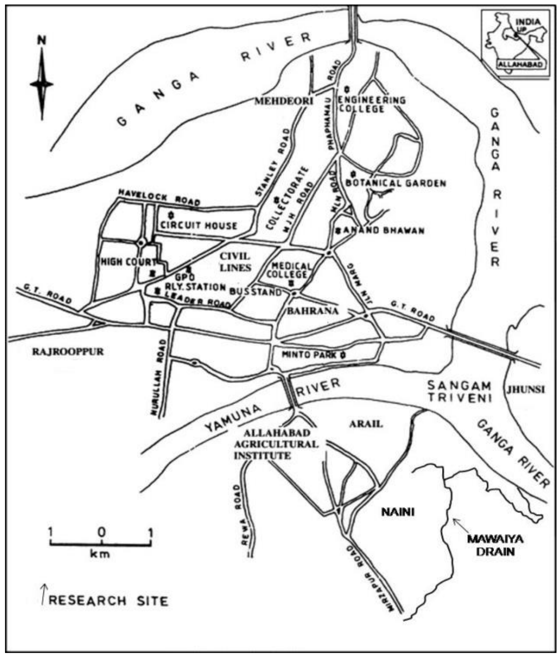

Figure.1 Mawaiya Drain of Naini, Allahabad.

Method of Study

Industrial wastewater samples of untreated and treated, top soil (0-13 cm. depths) and 10-15 plants of the same species of Tomato, Spinach, Coriandrum, Cabbage and Radish were collected randomly in triplicate by hand using vinyl gloves carefully packed into polyethylene bags and the whole plant body was brought to the laboratory from the suburban area of Naini Allahabad mawaiya drain during the month from December 2012 to June 2013 in order to estimate the total heavy metal content (Fe, Zn, Cd, Cu, Pb and Ni) in these samples. The cleaning (soil removal) of vegetable plant samples was performed by shaking and also by means of a dry pre-cleaned vinyl brush. Then the whole vegetable plant bodies were divided into different plants and non-edible portions were removed as per normal household practices. The edible parts of the vegetable samples were washed with tap water several times.

Wastewater samples were pretreated with concentrated HNO_3 to prevent Microbial degradation of heavy metals. 50 ml of wastewater samples were digested with 10 ml concentrated HNO_3 at 80^0C (APHA 1985). Soil samples were air-dried crushed and passed through 2 mm mesh sieve and stored at ambient temperature prior to analysis. The freshly harvested mature vegetables were brought to the laboratory and washed primarily with running tap water, followed by three consecutive washing with distilled water to remove the soil particles. Samples were cut into small pieces and dried in oven at 70^0C for 48 hour and then ground to powder 0.5 gm each of soil and vegetable samples were digested (wet acid digestion) with concentrated HNO_3, H_2SO_4 and $HClO_4$ (5:1:1) at 80^0C (Allen et al. 1986) until the solution become transparent. The digested samples of wastewater, 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 waste water, soil, and vegetable samples were estimated by atomic absorption spectrometer (Perkin Elmer analyst 400). Blank samples were analyzed after seven samples. Concentrations were calculated on a dry weight basis. All analysis was replicated three times. The accuracy and precision of metal analysis were checked against NIST-SRM, 1570 for every heavy metal. The result were found within ± 2 $^{0}/_{0}$ of heavy metals, mean, median, maximum, minimum, and standard deviation of wastewater, soil and vegetable samples were performed by using Microsoft Excel (version 2007).

III. RESULT AND DISCUSSION

Heavy metal status in wastewater

There are six elements examined in effluent-contaminated water used for irrigation in Allahabad industrial processing zone, the concentration of heavy metals in the untreated and treated sewage used for irrigation. In wastewater heavy metal concentrations (mg/L) ranged from 0.249 to 0.257 for Fe, 0.049 to 0.056 for Zn, 0.028 to 0.036 for Cd, 0.015 to 0.019 for Cu, 0.035 to 0.042 for Pb and 0.031 to 0.038 for Ni. In comparison with the standard guideline for irrigation water (Pescod 1992 and WHO 2007) it was found that mean value of Cd concentration in the wastewater exceeded the recommended level.

Table I: Heavy metal concentrations (mg/L) in wastewater used for irrigation in suburban area of Naini, Allahabad India (n = 27).

	Fe	Zn	Cd	Cu	Pb	Ni
Mean	0.253	0.052	0.032	0.017	0.038	0.034
Median	0.253	0.053	0.033	0.018	0.039	0.035
Minimum	0.249	0.049	0.028	0.015	0.035	0.031
Maximum	0.257	0.056	0.036	0.019	0.042	0.038
Std dev	0.004	0.003	0.004	0.002	0.003	0.003
Safe limit*	2.0	2.00	0.01	0.2	0.5	0.2

*Source: Pescod (1992) and WHO (2007).

Heavy metals concentration examined in which Fe was highest in wastewater used for irrigation in the study area. The mean concentration of Fe, Zn, Cd and Pb concentration in waste water of the study area were higher than the mean metal concentrations of irrigation water in suburban area of Varanasi, India (Sharma et al. 2007). The higher standard deviation observed for heavy metals of wastewater observe the highest Standard deviation in Cd and Fe in wastewater suggest that these metals were not uniformly distributed in the study area which is reported by Sharma et al. (2006). The data obtained from heavy metal in water from the present study varied more or less regularly with the findings of the other authors (Khan et al. 1998; Al Nakshabandi et al. 1997).

Heavy metal status in soil

The concentration of heavy metal in soil (mg/kg dry weight) in agricultural land of the study area (table II) ranged from 1345-1920 for Fe, 38.34-38.78 for Zn,31.23-31.24 for Cd, 32.54-35.26 for Cu, 18.21-18.32 for Pb and 117.2-117.6 for Ni. Besides Fe, the mean highest concentrations recorded in the soil was for Ni followed by Zn, Cu and Cd and the minimum concentration was observed for Pb which confirmed with the result reported by (Gupta et al.2008). Highest deposition of Fe in soil might be due to its long term use in the production of Machine tools, paints, pigments and alloying in various industries of the study area that may contaminate soil.

Table II**:** Concentration of heavy metals in wastewater irrigated soil (mg/kg dry soil) in suburban area of Allahabad, India (n = 49).

	Fe	Zn	Cd	Cu	Pb	Ni
Mean	1537	38.35	31.23	34.02	18.25	117.36
Median	1346	38.36	31.24	34.26	18.23	117.3
Minimum	1345	38.34	31.23	32.54	18.21	117.2
Maximum	1920	38.78	31.24	35.26	18.32	117.6
Std dev	331.6	0.744	0.005	1.375	0.058	0.208
Safe limit*	NA	300-600	3.0-6.0	135-270	250-500	75-150

*Source: Awashthi (2000) and WHO (2007).

The extent of metals observed in agricultural soil of the industrial area in the present investigation exceeded the permissible levels reported by different authors like Kabata - Pendias and Pendias (1992) (except Pb and Fe), Bowen (1966) (except Zn and Pb), and Temmerman et al. (1984) (except Pb and Fe). This variation of result might probably be due to the variations of heavy metals concentration in irrigation water and other agronomic practices of the respective area. The higher standard deviation reveals higher variations in the heavy metal distributions from the point source of emission to the adjacent areas. The low standard deviation of heavy metal in Cd may be described to its continuous removal by vegetables grown in the designated areas among the six heavy metals examined in soil and concentration of Fe was maximum (1920 mg/kg) with variation in its concentrations was several times higher than those of Kisku et al. (2000). In Naini Allahabad, the agricultural soil is contaminated with heavy metals through the repeated use of wastewater from industries and other sources in irrigation as well as application of chemical fertilizers and pesticides. Heavy metals occur in effluents of industries and in many fertilizers and pesticides. Cd, for example, is found in wastewater and also in phosphate fertilizers due to the presence of Cd as in impurity in all phosphate rocks. On many of agricultural soils, with the use of effluent contaminated water in irrigation, heavy doses of phosphate fertilizers have been applied for over 50 years, and all toxic trace elements found in these sources keep accumulating in soil (Alam et al. 2003).

Heavy metal status in vegetables

The average per capita consumption of leafy and non-leafy vegetables is 130gm/person/day (Alam et al. 2003). This is considerably less than the recommended amounts of 200gm/person/day from a nutritional point of view (Hasan and Ahmad 2000), the toxic trace elements concentrations in edible parts of vegetables grown in industrial area around Naini Allahabad are shown in figure 2 and 3. The range and mean concentration of trace metals (mg/kg dry weight) in leafy and other vegetables are presented in Table III. In leafy vegetables (Spinach, Cabbage and Coriander) the concentration of trace elements (mg/kg dry weight) ranged between 22.34-556.70 for Fe, 13.3-162.5 for Zn, 1.26-67.66 for Cd, 10.69-202.6 for Cu, 11.36-54.8 for Pb and 9.99-506.2 for Ni (Table III). The mean Fe concentrations varied from (32.49-351.23 mg/kg) which were good agreement with concentrations (111-378 mg/kg) observed in vegetables by Arora et al. (2008). The maximum uptake of Fe was in Tomato (351.23mg/kg) followed by Cabbage (292.35mg/kg) and Spinach (69.98 mg/kg), whereas the levels of Fe in all the vegetables were above the prescribed safe limit of WHO. The upper concentrations of Fe in vegetables were also found very similar to the values (2.11-336.9 mg/kg) reported by Kisku et al. (2000) for vegetables irrigated with mixed industrial effluents in India.

Table III: Heavy metal concentration (mg/kg dry weight) in vegetables grown in waste-water irrigated agricultural land in Allahabad: Mean and (Range).

Vegetables	Fe	Zn	Cd	Cu	Pb	Ni
Tomato (n=29)	351.23 (148.19-650.43)	28.23 (22.42-45.48)	2.36 (1.23-45.63)	23.12 (11.23-51.2)	12.20 (2.20-37.29)	10.11 (2.21-45.24)
Spinach (n=34)	69.98 (54.6-92.99)	36.50 (31.21-92.49)	15.24 (11.2-67.66)	29.20 (15.98-202.6)	16.20 (12.21-38.22)	66.55 (56.98-506.2)
Cabbage (n=26)	292.35 (130.25-556.70)	36.05 (23.54-79.05)	2.97 (1.26-3.92)	15.24 (10.69-26.97)	13.01 (11.36-35.96)	24.08 (9.99-39.35)
Coriander (n=48)	32.49 (22.34-45.32)	37.22 (13.3-162.5)	14.12 (7.65-29.36)	29.39 (17.23-38.26)	16.18 (12.6-54.8)	53.17 (52.9-72.8)
Radish (n=20)	45.26 (41.9-84.7)	34.7 (27.23-139)	18.92 (14.9-29.24)	32.20 (22.6-46.9)	17.26 (23.8-69.6)	58.82 (52.63-79.21)
Safe limit*	NA	50	1.5	30	2.5	1.5

*Source: (Awashthi 2000) and WHO (2007)

The highest mean concentration of Zn was found in Coriander (37.22 mg/kg) followed by Spinach (36.50 mg/kg). The mean concentration of Zn (28.23-37.22 mg/kg) in vegetables Allahabad industrial processing zone area of Naini was very similar to the vegetables from Beijing, China (32.01-69.26 mg/kg) (Liu et al. 2005), as also from Rajasthan, India (21.1-46.4 mg/kg) (Arora et al. 2008), but substantially lower than the Zn concentrations (3.00-171.03 mg/kg) in vegetables from Titagarh Waste Bengal,India (Gupta et al. 2008), Harare, Zimbabwe (1,038-1,872 mg/kg) (Thandi et al. 2004) and also the vegetables of Varanasi, India (59.61-79.46 mg/kg) (Sharma et al. 2008) and from Delhi, India (46.7-91.9 mg/kg) (Rattan et al. 2005). The maximum accumulation of Cd in Radish plants (18.92 mg/kg) followed by Spinach (15.24 mg/kg) and Coriander (14.12 mg/kg) which exceeded the WHO limit by approximately eight, five, and four times respectively. The present study revealed that the mean Cd level (2.36-18.92 mg/kg) measured in vegetables from Allahabad industrial processing zone area of Naini was higher than the vegetables from Titagarh West Bengal, India (10.37-17.79 mg/kg) (Gupta et al. 2008) and the vegetables from endemic upper gastrointestinal cancer region of Turkey (25 mg/kg) (Turkdogan et al. 2002). But lower than that the vegetables from China (0.03-0.73 mg/kg) (Liu et al. 2005) and significantly more than the vegetables from Egypt (0.002-0.08 mg/kg) (Doghien et al. 2004), where as it was very close to the findings of Sharma et al. (2007) (0.5-4.36 mg/kg) in vegetables from Varanasi, India.

In leafy vegetables (Spinach, Cabbage and coriander) the concentration of heavy metals (mg/kg dry weight) ranged between (22.34-556.70 mg/kg) for Fe, (13.3-162.5 mg/kg) for Zn, (1.26-67.66 mg/kg) for Cd, (10.69-202.6 mg/kg) for Cu, (11.36-54.8 mg/kg) for Pb and (9.99-506.2 mg/kg) for Ni (Table III). The mean Fe content in vegetables (32.49-351.23 mg/kg) was very higher to the result reported in Titagarh west Bengal, India (15.66-34.49 mg/kg) (Gupta et al. 2008) but lower than the Cu content in vegetables (61.20 mg/kg) from Zhengzhou city, China (Liu et al. 2006). However, the variation of Cu concentration in vegetable in the present study was strongly supported by the finding (5.21-18.2 mg/kg) of Arora et al. (2008) and also in good agreement with the values observed in Varanasi, India (10.95-28.58 mg/kg) by Sharma et al. (2007). Higher Cu concentration (202.6 mg/kg) was found in Spinach where as the mean value was (32.20-29.39 mg/kg) for Radish and Coriander, respectively which were lower than the mean value (32.74 mg/kg) and (36.41 mg/kg), respectively, reported by Sharma et al. (2008) in Varanasi India of the same vegetables. In addition Cu levels in vegetables showed good agreement with the main concentrations in leafy and non-leafy vegetables (15.5-8.51 mg/kg) from Samata Village, Jessor, Bangladesh obtained by Alam et al. (2003). The higher concentration of Cu level obtained in vegetables from present study than those of above authors, reveals the elevated uptake of the heavy metals in plants grown in industrial areas of Bangladesh.

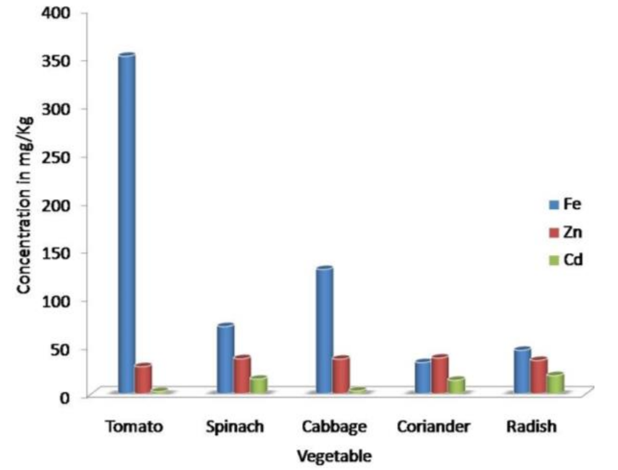

Figure 2: Mean heavy metal concentration (Fe, Zn and Cd) in vegetables of industrial area of Naini Allahabad.

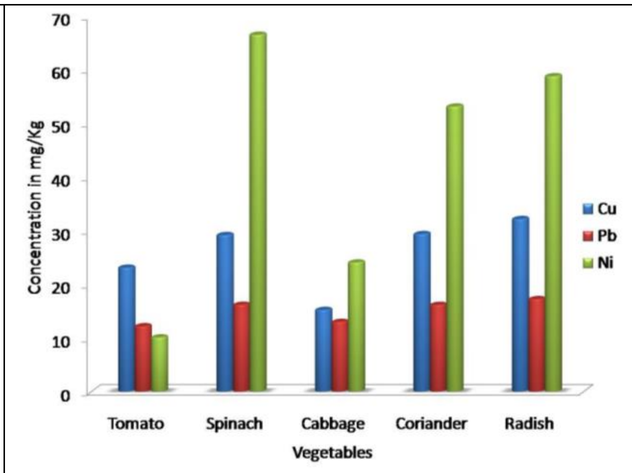

Figure 3: Mean heavy metal concentration (Cu, Pb, Ni,) in vegetables of industrial area of Naini Allahabad.

The maximum concentration of Pb was exhibited by Radish (17.26 mg/kg) followed by Spinach (16.20 mg/kg) which exceeded the acceptable tolerance level of WHO for Pb by three and three times, respectively. Pb concentrations in edible portions of all the vegetables examined in the present study were above the permissible levels recommended by WHO, India (Awashthi 2000) The mean Pb content in vegetables(12.20-17.26 mg/kg) was lower than the values reported in Titagarh, West Bengal, (21.59-57.63 mg/kg) (Gupta et al. 2008) but comparatively higher than the Pb level reported in China (0.18-7.75 mg/kg) (Liu et al. 2006), (1.97-3.81 mg/kg) (Liu et al.2005) and in Varansi, India (3.09-15.74 mg/kg) (Sharma et al.2007). However it was significantly lower than the mean concentration of Pb (409 mg/kg) reported in vegetables from Turkey by Turkdogan et al. (2002). Higher concentration of Ni shown by Spinach (66.55 mg/kg) was this fold higher than the recommended safe limit of PFA (Prevention of food adulteration) (Awashthi 2000). The mean Ni concentrations in vegetables varied from (10.11-66.55 mg/kg) which was lower than the data reported by vegetables in Titagarh, West Bengal, India by Gupta et al. (2008). However, it was higher to the findings of Sharma et al. (2007) (1.81-7.57 mg/kg) in Varanasi, India Rattan et al. (2005) (8.78-21.5 mg/kg) in Delhi, India.

Bioconcentration factor

Bioconcentration factor or plant concentration factor (PCF) is a parameter used to describe the transfer of trace elements from soil to plant Edible parts. It is calculated as the ratio between the concentration of heavy metals in the vegetables and that in the corresponding soil all based on (dry weight) for each vegetable separately (Liu et al. 2006).

Bioconcentration factor or Plant concentration factor = C_{plant} / C_{soil}

Where C_{plant} and C_{soil} represents the heavy metal concentration in extracts of plants and soils on a dry weight basis respectively.

Table IV: Bioconcentration Factor/Plant concentration factor of the study area.

Vegetables	Fe	Zn	Cd	Cu	Pb	Ni
Tomato (n=29)	0.228	0.727	0.075	0.679	0.668	0.086
Spinach (n=34)	0.045	0.941	0.487	0.858	0.887	0.567
Cabbage (n=26)	0.190	0.929	0.095	0.447	0.712	0.205
Coriander (n=48)	0.021	0.959	0.452	0.863	0.886	0.453
Radish (n=20)	0.029	0.894	0.605	0.946	0.945	0.501

The BCF OR PCF value ranges from Fe, 0.021-0.228, Zn 0.727-0.959, Cd 0.075-0.605, Cu 0.447-0.946, Pb 0.668-0.945 and Ni 0.086-0.567. The BCF Value for Zn (0.959) of Coriander was the highest among all considered heavy metals which was supported by the observation of Liu et al. (2006). The trends of BCF For heavy metals in different vegetables studied were in the order of Zn > Cu > Pb > Cd > Ni > Fe, were as more or less similar to result reported by Khan et al. (2008) in Beijing, China. Although Zn concentration in soils of the study area were below the threshold levels of WHO and other authors, the highest mean BCF value of Zn in vegetables might be attributed to the exceeding high concentration of Zn, is (0.052 mg/L), in irrigation water of the study area (Table I).

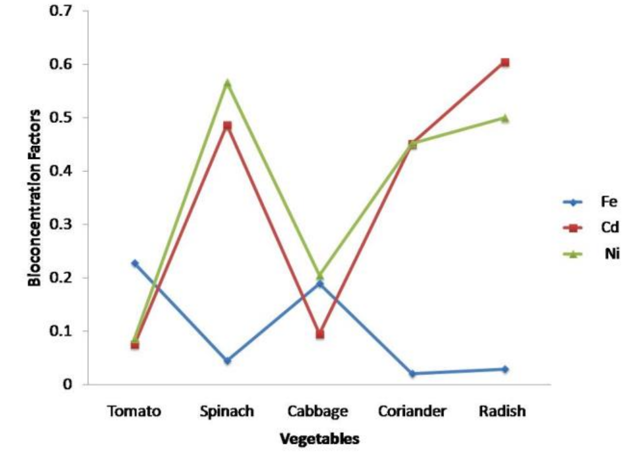

Figure 4: Bioconcentration Factors of Fe, Cd and Ni in different vegetables

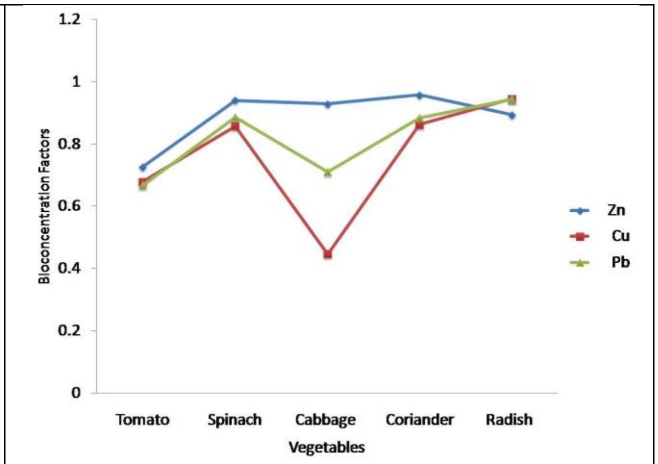

Figure 5: Bioconcentration Factors of Zn, Cu and Pb in different vegetables

Moreover the exchangeable Zn, which was considered readily bioavailable, accounted for ≤ 10% of the total Zn in Crop soils in general (Wong et al. 2002). Therefore, enrichment of vegetable for Zn might also be another pathway except absorption from soils. Of the five vegetable species, Tomato (for Zn and Cu), Spinach (for Ni), Cabbage (for Fe), Coriander (for Cd) and Radish (for Pb) showed highest metal accumulation. Vegetables differed in their ability to accumulate and concentrate metals in their edible parts, differences between them were significant which was well supported from the studies carried out by Sharma et al. (2006). The variations in heavy metal concentrations in vegetables were due to variations in their absorption and accumulation tendency. Soil properties such as Ph, organic matter, cation exchange capacity, redox botanical, Soil texture, and clay content may also affect the heavy metal uptake (Overesch et al. 2007). The interaction between different heavy metals occurs at root surface and also within the plant, which ultimately affects the uptake and translocation of heavy metals (Sharma et al. 2006). The accumulation of heavy metals also depends on plant age and plants parts (Liu et al. 2007).

IV. CONCLUSION

From the results of the present study it is clear that and elevated level of metal accumulation in edible parts of vegetable plants is mainly from their growth habitat like water and soil in and around industrial areas of Naini Allahabad. Long term consumption of these metal-contaminated vegetables can cause different disease like Brain and Kidney damage, cancer in human body, dermatitis. The data generated must be used as baseline wastewater quality framework to serve as a basis for monitoring irrigation water quality in rurban areas of Allahabad to ensure safety. Responsible agencies should carry out public health education within the consumption area to sensitive the general public on the potential effects of indiscriminate disposal of waste and the potential health hazards associated with the consumption of vegetables cultivated with wastewater. Measures must be taken to reduce heavy metal pollution and nutrient loading of irrigation water and soils to protect the safety of both farmers and consumers.

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