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Assessment of Bio-Concentration Factor of Heavy Metals in Indian Soil-Crop System

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Bioaccumulation of heavy metals in different agricultural products in soil-crop systems has fascinated pervasive attention in yester years due to food safety issues. The objectives of this study were to determine the influence of different soil parameters on bio-concentration of heavy metals' from soil to pearl millet grains. For the purpose, heavy metals were quantified in pearl millet grains and corresponding fields' rhizospheric soil samples from different study sites in Haryana, India. The mean concentrations of Cd, Pb, Ni, Zn, Fe and Cu in pearl miller garins were found to be 0.50, 6.74, 3.86, 49.72, 142.0 and 10.17 mg kg^{-1} respectively. Pearl millet showed capacity to transfer essential metals Zn and Cu from soil to grains while showed a restricting effect to the uptake of Co and Cr. Further multivariate statistical techniques like correlation analysis, principal component analysis and cluster analysis were applied to the data for supplementary and qualitative evaluation of inter-dependences among the studied parameters. Bio-concentration factors of all the studied heavy metals in pearl millet grains showed significant negative correlations with their respective soils' total metal concentrations. The results revealed lesser metal uptake with increasing soil total metal concentrations. Inspite of the variability in the characteristic of investigated soils, significant correlations among soil quality parameters and metal accumulation in grains were recorded. Cluster analysis revealed formation of many primary cluster pairs such as EC- Na, CEC- Ca, BCF_{Ni} - Cu_s, BCF_{Fe} - TOC, BCF_{zn} - EC, BCF_{Pb} - pH, BCF_{Cu} - Na and Cd_S- Pb_s etc.

Keywords: Heavy Metals, Pearl Millet, Bio-Concentration Factor, Correlation, Principal Component Analysis

Introduction

Heavy metals in the environment might occur naturally from parent materials, forest fires wind blown dusts, volcanic eruptions and marine aerosols[1]. Various anthropogenic activities includes excessive use of agrochemicals, sewage water irrigation, combustion of petroleum in automobiles and coal products in energy production, metal tailings, smelting, refining and transportation in mining and smelting industries, incineration and fly ash also release metals and metals laden wastes in the environment[1,2]. With the rapid growth in industrialization in past few decades, soil contamination with heavy metals has emerged as a global environmental problem[3]. Due to non-degradable and persistent nature heavy metals get accumulated in soils to levels, interfering their natural ability to limit the toxicity and metals may be mobilized, contaminating groundwater, agricultural products and other environmental media[4,5]. It is reported that heavy metals may enter the food chain depending individual heavy metals' reactions with soils and translocation rates in plants, thus of ecological, nutritional and environmental significance[6]. Pearl millet (*Pennisetum glaucum*) is the most widely grown type of millet cultivated over 260,000 km^2 worldwide and accounts for approximately 50% of the total world production of millets[7]. The present study focused on heavy metals' quantification of crop - soil system and to assess the influence of various soil quality parameters on the bio-concentration factors (BCF) of heavy metals from soil to pearl millet grains.

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Experimental Section

Study Area

Haryana state is located in Northwest of India between a latitude of 30.30° North and longitude of 74.60° East sharing its boundaries with Rajasthan in south and west, Himachal Pradesh and Punjab in the north, and the territory of Delhi in the east. The study area is an alluvial plain of Indo-Gangetic basin having sandy, sandy loam and clay soil types and located in south-western part of state. With flat plain topography this region has an average elevation 208 m gently sloping from north-east to south-west[8]. Climate is of tropical type with wide variation in temperature from 47 °C in summer to 2 °C in winter. The area receives an annual rainfall of 395.6 mm, 71% of which is received during south-west monsoon period, July to September.

Sample Collection and Processing

Pearl millet grains sampling and analysis

Pearl millet grain samples from 23 sampling locations during its harvesting season were collected, sealed in plastic containers and brought to the laboratory for further analysis. The grain samples were dried, ashed and homogenised and then 0.2 g of each ash samples was digested in a microwave digester (CEM Mars X) using 5.0 ml diacid (HNO_3 and $HClO_4$) mixture. Digests were quantitatively transferred into glass beakers after cooling digestion vessels at room temperature. After evaporating the digests near dryness on hot plate, residues were dissolved in double distilled water to make desired volume. The prepared samples were refrigerated until analysis in polypropylene bottles. In flame atomic absorption spectrophotometer (FAAS) acetylene gas was used as fuel and air as support and an oxidising flame was used in all the cases except chromium, where reducing nitrous oxide flame was used to quantify metal. The detection limits of AAS for Fe, Cu, Cd, Ni, Zn, Cr, Co and Pb are 0.45, 0.025, 0.01, 0.04, 0.01, 0.05, 0.05 and 0.06 mg L^{-1} respectively. The heavy metal concentrations were finally expressed in mg kg^{-1} on dry wt. basis. To ensure the reliability of results, standards of respective metals were run after every 10 samples analyzed.

Soil sampling and analysis

Corresponding sub surface rhizospheric soil samples were collected from pearl millet fields'. From rectangular grid of 1.0 m^2, four sub-samples of soil from a depth of 5-10 cm were collected and mixed together to obtain a representative sample. Care was taken to get rid of the metal contamination, using non metallic spade while collecting soil samples. Samples were sealed in clean plastic bags after removing foreign bodies and taken to laboratory. Soil samples were air dried, grounded and then passed through a 2 mm sieve. Like grain samples, soil samples were digested and analyzed for their heavy metals contents.

Statistical Analysis

Statistical techniques like correlation analysis, Principal component analysis and Cluster analysis were applied to the data using SPSS software package (version 16.0).

Bio-concentration factor of heavy metals from soil to pearl millet grains

Soils to pearl millet grain BCFs were calculated to evaluate the uptake efficiency order of heavy metals' by the grains in the study area. It is computed as the ratio of concentration of the heavy metal in grains to their concentration in respective soils[3].

$$BCF = \frac{C_{grain}}{C_{Soil}} \quad \text{... (1)}$$

Where, C_{grain} and C_{soil} are the concentration of heavy metal in grain and rooted soils on dry weight (DW) basis, respectively.

Results and Discussion

Heavy metals concentration in pearl millet fields soils

Pearl millet field's rhizospheric soils' total heavy metal concentrations on dry weight (DW) basis along with their physic-chemical properties are given in Table 1. Iron concentration in soil samples varied from 1584 - 6297 mg kg^{-1} followed by Zn (71.04 - 175.20 mg kg^{-1}) > Pb (29.99 -148.24 mg kg^{-1}) > Cu (16.41 -38.16 mg kg^{-1}) > Ni (9.60 -32.86 mg kg^{-1}) > Cr (1.10 -9.0 mg kg^{-1}) > Cd (0.62 -8.47 mg kg^{-1}) > Co (1.03 -6.24 mg kg^{-1}). Mean concentrations (mg kg^{-1}) of Cu (25.95), Cd (3.69), Pb (66.56), Ni (18.96) and Zn (115.32) were well within permissible limits for Cu (135 - 270 mg kg^{-1}), Cd (3 - 6 mg kg^{-1}), Pb (250 - 500 mg kg^{-1}), Ni (75-150 mg kg^{-1}) and Zn (300 - 600 mg kg^{-1}) with reference to Indian standards[9]. In the present study mean concentrations of Cd, Zn, Pb, Ni and Cu were higher than mean concentrations of (2.80, 20.35, 15.57, 43.56, 13.37 and 30.67 mg kg^{-1}) for Cd, Cu, Pb, Zn and Ni as reported in the soils of wastewater irrigated area of Dinapur[10]. Soil properties namely pH, texture, moisture content, organic matter and nutrient status along with plant part concerned, age and nature of metallic species may influence heavy metals uptake[3]. Therefore different soil quality parameters were analyzed and the same are given in Table 1. To identify any relationships between heavy metals concentrations of fields' soils and its quality parameters, Pearson's correlation analysis was performed. Significant positive correlation was found between Cd and Pb content in soil (r = 0.70**; P<0.01) suggesting their same anthropogenic origin as both the metals are human induced. Soil total Zn was found significantly positively correlated with EC (r = 0.44*; P<0.05). However physico-chemical parameters correlation matrices revealed positive correlations among them such as EC - Na (r = 0.70**; P<0.01), Na- Ca (r = 0.44*; P<0.05), Ca- CEC (r = 0.47*; P<0.05) and CEC - K (r = 0.43*; P<0.05). Total organic carbon (TOC) was found negatively correlated with pH (r = - 0.88**; P< 0.01). Decrease in pH with increasing TOC contents of the soil may be attributed due to formation of more organic acids, thus lowering the pH of the soil solution.

Heavy metals and their bio-concentration in pearl millet grains

Heavy metals' concentration and their respective BCF from soil to pearl millet grains collected from study site are given in Table 2. Average Cd, Pb, Ni, Zn, Fe and Cu concentrations in pearl millet grain were found to be 0.50, 6.74, 3.86 49.72, 142.0 and 10.17 mg kg^{-1} respectively, however Co and Cr were found to be below detection limit (BDL). In collected grain samples essential metals Fe, Cu and Zn content was much higher than the other metals under study. Adverse non-carcinogenic health impacts posed by toxic heavy metals include hepatic, renal, neurological, developmental, gastrointestinal, respiratory, cardiovascular, hematological, reproductive, immunological disorders etc. The mobility of the heavy metals from rhizospheric soils to the harvestable aerial regenerative parts of the plant i.e. grains was evaluated as BCF and given in Table 2. As Co and Cr content in collected grain samples were BDL therefore no BCF values for these respective metals could not be calculated. The mean BCFs of heavy metals from soil to pearl millet grains were in the order of Zn > Cu >Ni > Cd > Pb > Fe. Zinc BCF with a mean value of 0.45 was higher as compared to other studied heavy metals while that of iron was minimum with a mean value of 0.06. In the present study a wide variation in BCF values of heavy metals at different locations of the study area was observed (Table 2). This may be attributed to variation in the soil characterstics as well as the sources and levels of

Table 1 — Total heavy metals' concentration (mg kg^{-1}) and different soil quality parameters of pearl millet fields under study

Metal	Range (mg kg^{-1})		Mean	± S. D.	Soil quality parameters	Range		Mean	± S. D.
	Min	Max				Min	Max		
Fe	1584	6297	2708	± 1242	pH	7.1	7.9	7.60	± 0.22
Cu	16.41	38.16	25.95	± 6.15	EC (µs cm^{-1})	60.98	470.80	128.69	± 90.18
Cd	0.62	8.47	3.69	± 2.44	Na (mg kg^{-1})	212	1086	397.26	±192.66
Pb	29.99	148.24	66.56	± 32.0	K (mg kg^{-1})	222	650	411.52	± 121.0
Ni	9.60	32.86	18.96	± 5.31	CEC(meq100g^{-1})	53.85	108.86	81.79	± 12.51
Zn	71.04	175.20	115.32	± 30.19	TOC (%)	0.65	1.91	1.09	± 0.35
Co	1.03	6.24	3.48	± 1.65	Ca (g kg^{-1})	0.45	3.55	0.96	± 0.60
Cr	1.10	9.00	4.50	± 2.30					

Table 2 -— Heavy metals' content (mg kg^{-1}) and their respective bio- concentration factors from soil to pearl millet grains

Metals	Pearl millet grains (mg kg^{-1})			Bio concentration Factor		
	Range	Mean	± S. D.	Range	Mean	± S. D.
Fe	39.7 - 324.7	142.00	± 63.17	0.008 - 0.135	0.060	± 0.029
Cu	3.43 - 16.91	10.17	± 4.32	0.112 - 0.805	0.423	±0.224
Cd	0.13 - 1.24	0.50	± 0.29	0.043 - 0.493	0.176	± 0.107
Pb	2.35 - 14.70	6.74	± 3.84	0.022 - 0.265	0.111	± 0.064
Ni	[a]BDL - 7.71	3.86	± 1.65	BDL - 0.586	0.223	± 0.128
Zn	13.74 - 89.65	49.72	± 22.08	0.126 - 0.911	0.450	± 0.209
Co	[b]BDL	-	-	-	-	-
Cr	[c]BDL	-	-	-	-	-

[a]BDL -Below detectable limits for Ni (<0.04 ppm)
[b]BDL -Below detectable limit for Co (<0.05 ppm)
[c]BDL -Below detectable limit for Cr (<0.05 ppm)

metals present in different soils. Significant negative correlations at $P<0.01$ were reported between the pairs like BCF_{Fe} - Fe_s (r = -0.58), BCF_{Cu} - Cu_s (r = -0.61), BCF_{Cd} - Cds (r = - 0.59), and BCF_{Ni} - Ni_s (r = -0.59) revealing lesser metal uptake in grains at higher metal concentration in soil of the respective agricultural fields'. Similar observations have been reported by Lobben (1993)[10] who found lesser heavy metals transfer for various crop species on contaminated soils compared to uncontaminated control sites. Soil organic matter enhanced iron uptake by pearl millet as BCF_{Fe} showed positive correlation with TOC (r = 0.38**; $P<0.01$) suggesting the presence of higher bio-available fraction of Fe in soil solution at higher TOC. Higher the TOC content of the soil, more will be the retention of different ions on its surface against the leaching, thus greater the uptake by plants. On the contrary, significant negative correlation between Cd contents of pearl millet grain and soil organic matter was reported by Kramer and Konig (1982)[11]. Further the metal bio-concentration coefficient showed negative correlation with pH (r = -0.44*; $P<0.05$). Lesser the soil pH more the no. of H^+ ions that can be released in soil solution favouring the transfer ability of heavy metals from soil to plant facilitated through H^+-coupled transport[12,13]. Uptake of Zn from soil solution was found positively correlated with EC (r = 0.44*; $P<0.05$) and that of Ni was found correlated negatively with K (r = - 0.46*; $P<0.05$).

Principal component analysis and Cluster analysis

Results of correlation matrix suggested several significant correlations whether positive or negative among BCF of heavy metals in pearl millet grains, soil total metal concentration in soil and its physico-chemical parameters and were supported by dendrogram of cluster analysis (Fig. 1). The clustering behaviour showed primary cluster pairs between EC - Na, CEC - Ca, BCF_{Ni} - Cu_s, BCF_{Fe} - TOC, BCF_{Zn} - EC, BCF_{Pb} - pH, BCF_{Cu} - Na and Cd_S - Pb_s. Principal component analysis with Varimax normalization (PCA-V) was applied for the qualitative evaluation of clustering behaviours and mainly five factors explaining a total variance of 63 % were obtained. Factor-1 contributed 15.64 % to the total variance with high loading on various soil quality parameters and soil heavy metals; K (r = 0.60), CEC (r = 0.42), Cd_S (r = 0.87) and Pb_s (r = 0.78) thus explaining K - CEC and Cd_s - Pb_s cluster pairs. Factor-2 contributed

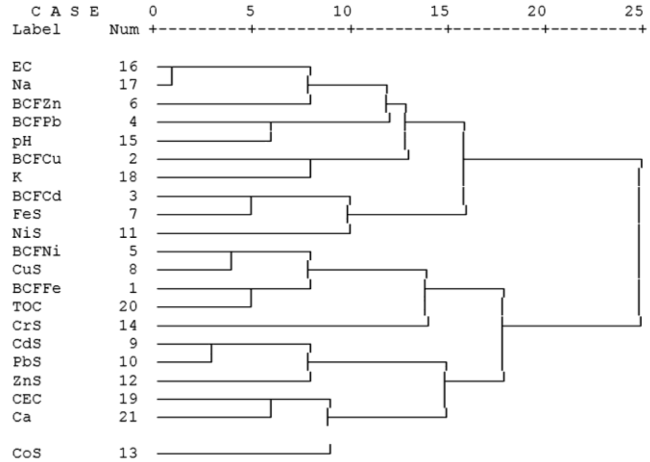

Fig. 1 — Dendrogram for selected heavy metals in soil, their bio-concentration factor in pearl millet grains and soil physico-chemical parameters.

13.11% to the total variance with high loading on BCF_{Pb} (r = 0.50) and pH (r = 0.95) thus explaining BCF_{Pb} - pH cluster pair. Factor-3 contributed 12.53% to the total variance with high loadings on soil quality parameters Na (r = 0.89), EC (r = 0.93), K (r = 0.53), Ca (r = 0.47), CEC (r = 0.42) and BCF_{Zn} (r = 0.43) thus explaining EC-Na, CEC - Ca and BCF_{Zn} - EC cluster pairs observed in cluster analysis dendrogram. Factor-4 with higher loadings on BCF_{Ni} (r = 0.79), Cu_s (r = 0.58), BCF_{Fe} (r = 0.52) and TOC (r = 0.21) explained Ni_{BCF} - Cu_s and BCF_{Fe} - TOC pairs and contributed 11.87 % to the total variance. Factor-5 with a variance of 9.5 % with high loadings on BCF_{Zn} (r = 0.95) and K (r = 0.53) supported BCF_{Zn} - K cluster pair.

Conclusion

Soil pH, TOC, EC and K were the significant variables controlling heavy metals' accumulation in pearl millet grains. Study elucidated negative associations of BCF of heavy metals in pearl millet grains with that of respective fields' soil total heavy metal contents, concluding decreased metal uptake by grains with increasing soil metal concentrations. A wide variation in BCF of heavy metals was observed which might be accounted to different sources and levels of contaminants present in different soils coupled with variable soil quality parameters.

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