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Heavy metals distribution and their correlation with physico-chemical properties of different soil series of northwestern India

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ABSTRACT

A study on "spatial distribution of heavy metals content and physico-chemical properties of different soil series of northwestern India" was conducted during 2017-18 in five well established soil series which are under aeolian and upper alluvial plain of Hisar district (Haryana). The 50 surface (0-15cm) soil samples were collected with the help of GPS system (*Global Positioning System)* and analyzed for physico-chemical properties and availability of heavy metals (Pb, Cr, Cd, Co and Ni). The soil of different soil series of Hisar district were moderately alkaline to strongly alkaline in nature (pH 8.0-8.5), electrical conductivity (EC) ranged from 0.10-0.17 dS m^{-1} (non saline).The organic carbon content in soil of different soil series was low to medium (0.39-0.51%) and having sandy to sandy clay loam texture with low to moderate CEC (8.64-12.85 cmol kg^{-1}). The calcium carbonate content in soils was < 5 %. The mean of available heavy metals such as Pb, Cr, Cd, Co and Ni ranged from 0.56-1.30, 0.03-0.17, 0.01-0.04, 0.00-0.02 and 0.03-0.16 mg kg^{-1} respectively, which were found below the permissible limit.

Key words: Field survey, Heavy metals, Soil series, Spatial distribution

Heavy metals are large group of elements with density, generally greater than 5 g/cm^3. Due to non-degradable nature, buildup of heavy metals in soil takes place. In due course of time these metals enter into the food chain which impart negative impact on plants (e.g. photosynthesis, gaseous exchange, nutrient absorption, reductions in growth and dry matter accumulation *etc.*), animals and human beings (Zhu *et al.* 2018). In small concentrations, the heavy metals in plants or animals are non toxic but lead (Pb) and cadmium (Cd) are exceptions which are always toxic even in very low concentrations (Sun *et al.* 2019). Some other metals such as copper (Cu), cadmium (Cd), zinc (Zn), manganese (Mn), cobalt (Co) and molybdenum (Mo) acts as micronutrients for the growth of plant, whereas others such as Cd, As and Cr acts as carcinogens (Vodyanitskii 2016) and Hg and Pb are associated with the development of abnormalities in children (Mensah *et al.* 2017).

Due to agro-climatic conditions, anthropogenic activities, presence and distribution of heavy metals are inevitable (Hua *et al.* 2018). Soils contain these metals in the form of inorganic compounds or they may remain bonded with organic matter, clays or as oxides (Shaheen and Iqbal 2018). Heavy metals in soil may exist in one or more following forms: (a) as dissolved (in soil solution), (b) as exchangeable (in inorganic components), (c) as structural components of the lattices of soil minerals, (d) as insoluble precipitates with other soil components (Wang *et al.* 2019). These metals have discriminative characteristics such as (i) they don't decay with time, (ii) they are beneficial to plants at certain permissible limits but can be toxic when exceeding specific thresholds levels, (iii) they present at a background level of non-anthropogenic origin and (iv) they strongly interact with the soil matrix, consequently, heavy metals in soils can become mobile as a result of changing environmental conditions (Zhang *et al.* 2018). The distribution of heavy metals in solid phases depends on their amounts, their type and origin (Roca 2015). The binding mechanisms of heavy metals vary with soil depth, depending on the physical and chemical properties of the soil such as pH, redox potential and water content, all these play an important role for the rate of chemical transformation of an element in a solid phase (Khan *et al.* 2016). The accumulation of heavy metals in soils of agricultural field and lands is of increasing concern due to food safety, potential health risks as well as its deleterious

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effects on floral and faunal soil ecosystems (Qishlaqi and Moore 2007).

Therefore, their assimilation in soil is very likely. Hence, it may create a health alarm to all farmers as well as the consumers of different food crops/fruits/vegetables those are being consumed directly to meet the food needs. Such studies are very useful to estimate the level of heavy metal contamination and risk in agricultural soils to assure crop quality and soil health (Chahal *et al.* 2014). Such type of research work has not been carried out in soil series of Hisar district (Haryana). Thus, keeping in view the above facts, the present study was carried out.

MATERIALS AND METHODS

The present study of heavy metals status in different soil series was carried out in Hisar division of Haryana in northwestern India. To find out the heavy metal content in different soil series of Hisar district, this study was conducted in a part of the Indo-Gangetic alluvial plain, covering an area of 6560 square km, which lies between 28.59° to 29.49° N latitude and 75.11° to 76.18° longitude. The GPS based surface (0-15 cm) soil samples were collected randomly from the five well established soil series . Soil series of Balsamand, Isarwal and Rawalwas located at latitude 29°05'293" N, 29°06'393" N, 29°08'716" N and longitude 75°44'560" E, 75°46'436" E, 75°58'162" E, respectively which becomes under aeolian plain. Barwala and Ladwa series located at latitude 29°23'389" N, 29°15'036" N and longitude 75°90'884" E, 75°90'884" E, respectively and becomes under alluvial plain. Soil samples were dried at room temperature and ground on wooden pestle and mortar and passed through 2 mm sieve. The soil samples were analyzed for soil reaction (pH) and electrical conductivity (EC) in a soil: water suspension of 1:2 ratios by (Jackson 1973). Organic carbon (OC) content was determined by wet oxidation method (Walkley and Black 1934), calcium carbonate by rapid titration method (Puri 1949), soil texture by International pipette method (Piper 1950) and cation exchange capacity (CEC) by ammonium acetate method (Hesse 1971). DTPA extractable Pb, Cr, Cd, Co and Ni contents were determined in soil by using AAS (Model-Z-Xpress 8000) as per procedure given by Lindsay and Norvell (1978).

RESULTS AND DISCUSSION

Physico-chemical properties of soils

The data on soil pH of surface soil samples (0-15 cm) of Balsamand soil series ranged from 8.0 to 8.4 with a mean of 8.2, Isarwal soil series ranged from 7.3 to 8.5 with a mean of 8.0, Rawalwas soil series ranged from 8.0 to 8.4 with a mean of 8.1, Barwala soil series ranged from 8.0 to 8.7 with a mean of 8.3 and in Ladwa soil series it varied from 8.3 to 8.7 with a mean of 8.5 (Table 1). The soil reaction of study area was found under moderately alkaline (8.2) in Balsamand, Isarwal, Rawalwas and Barwala soil series but strongly alkaline (8.5) in Ladwa soil series. The overall mean range of the pH varied from 8.0 to 8.5. Urmila *et al.* (2015) also reported similar results in terms of soil pH of Rohtak (Haryana), which are associated with present study. The pH of the soil controls the availability of metals. The solubility of metal complexes increased with decrease in soil pH which shows, negative correlation between Cu, Pb, Cd and Ni with pH. The ability of soils to increase the immobilization of heavy metals with rising pH in the present study. The concentration of soluble salts (EC) of Balsamand soil series ranged from 0.07 to 0.16 dS m^{-1} with a mean of 0.11 dS m^{-1}, Isarwal soil series ranged from 0.03 to 0.25 dS m^{-1} with a mean of 0.10 dS m^{-1}, Rawalwas soil series ranged from 0.05 to 0.14 dS m^{-1} with a mean of 0.14 dS m^{-1}, Barwala soil series ranged from 0.10 to 0.28 dS m^{-1} with a mean of 0.16 dS m^{-1} and in Ladwa soil series it varied from 0.10 to 0.38 dS m^{-1} with a mean of 0.17 dS m^{-1}. The overall mean range of the electrical conductivity varied from 0.10 to 0.17 dS m^{-1}. Results of present study are closely associated with Sharma and Prasad (2010).

The organic carbon (%) of different soil series of Haryana was found low in Balsamand, Isarwal and Rawalwas but it was medium in Barwala and Ladwa soil series. The organic carbon content of Balsamand soil series

Table 1 Physico-chemical properties of different soil series in surface layer (0-15 cm)

Soil series	pH (1:2)	EC (dS m^{-1})	Organic carbon (%)	CEC (cmol kg^{-1})	$CaCO_3$ (%)	Sand (%)	Silt (%)	Clay (%)
Balsamand	8.0-8.4 (8.2)*	0.07-0.16 (0.11)	0.25-0.47 (0.39)	7.53-10.16 (8.64)	2.50-3.25 (2.95)	85-92 (89)	2-9 (4.6)	5-8 (6.4)
Isarwal	7.3-8.5 (8.0)	0.03-0.25 (0.10)	0.21-0.59 (0.43)	6.91-10.42 (8.77)	2.25-3.25 (3.00)	85-92 (84.5)	2-11 (7.1)	5-15 (8.3)
Rawalwas	8.0-8.4 (8.1)	0.05-0.14 (0.14)	0.29-0.59 (0.47)	7.53-10.16 (9.13)	3.00-4.50 (3.92)	70-86 (79.7)	2-16 (7.5)	7-19 (12.8)
Barwala	8.0-8.7 (8.3)	0.10-0.28 (0.16)	0.43-0.60 (0.51)	10.97-15.38 (12.85)	3.00-3.50 (3.13)	46-80 (59.1)	2-29 (20.8)	15-24 (20.1)
Ladwa	8.3-8.7 (8.5)	0.10-0.38 (0.17)	0.33-0.60 (0.48)	9.39-15.38 (11.73)	2.75-3.50 (3.25)	45-79 (59.1)	3-31 (20.2)	17-24 (20.7)

*Parentheses - Mean value

ranged from 0.25 to 0.47% with a mean of 0.39%, Isarwal soil series ranged from 0.21 to 0.59% with a mean of 0.43%, Rawalwas soil series ranged from 0.29 to 0.59% with a mean of 0.47%, Barwala soil series ranged from 0.43 to 0.60% with a mean of 0.51% and in Ladwa soil series it ranged from 0.33 to 0.60% with a mean of 0.48%. Organic matter content, determine the soil's ability to retain and immobilize the heavy metals. Organic matter plays an important role not only in forming complexes but also in retaining heavy metals in an exchangeable form. The overall mean range of the organic carbon varied from 0.39 to 0.51%. Qishlaqi and Moore (2007) also reported similar results in soils of Iran. The data on calcium carbonate of Balsamand soil series ranged from 2.50 to 3.25% with a mean value of 2.95% (Table 1), Isarwal soil series ranged from 2.25 to 3.25% with a mean value of 3.00%. In Rawalwas soil series, $CaCO_3$ ranged from 3.00 to 4.50% with a mean value of 3.92%. In case of Barwala soil series, $CaCO_3$ ranged from 3.00 to 3.50% with a mean value of 3.13%. It ranged from 2.75 to 3.50% in Ladwa soil series with a mean value of 3.25 %. The heavy metals concentration generally decreased with depth associated with increasing pH and $CaCO_3$ contents that put forth a major influence on the availability heavy metals. Yerima *et al.* (2013) also reported the similar results. The data on sand, silt and clay ranged from 85 to 92, 2 to 9 and 5 to 8% with a mean value of 89, 4.6 and 6.4 %, respectively in Balsamand soil series. In Isarwal soil series the sand, silt and clay ranged from 85 to 92, 2 to 11 and 5 to 15% with a mean value of 84.5, 7.1 and 8.3 %, respectively. In Rawalwas soil series the sand, silt and clay ranged from 70 to 86, 2 to 16 and 7 to 19 % with a mean value of 79.7, 7.5 and 12.8%, respectively. The sand, silt and clay ranged from 46 to 80, 2 to 29 and 15 to 24% with a mean value of 59.1, 20.8 and 20.1%, respectively in Barwala soil series (Table 1) and in Ladwa soil series the sand, silt and clay ranged from 45 to 79, 3 to 31 and 17 to 24% with a mean value of 59.1, 20.2 and 20.7%, respectively. The soil texture of study area was found sandy containing 89, 4.6 and 6.4% sand, silt and clay, respectively in Balsamand soil series, loamy sand (84.5, 7.1 and 8.3 %) in Isarwal soil series, loamy sand (79.7, 7.5 and 12.8 %) in Rawalwas soil series, sandy clay loam (59.1, 20.8 and 20.1 %) in Barwala soil series and sandy clay loam (59.1, 20.2 and 20.7 %) in Ladwa soil series. The levels of heavy metals in the present studied soils were much below the permissible limits due to leaching of metals to lower soil layers, sandy texture and precipitation during *kharif* season. Bhatti *et al.* (2015) also reported the similar results in soils of Punjab.

The CEC of Balsamand soil series ranged from 7.53 to 10.16 cmol kg^{-1} with a mean of 8.64 cmol kg^{-1}, Isarwal soil series ranged from 6.91 to 10.42 cmol kg^{-1} with a mean of 8.77 cmol kg^{-1}. In Rawalwas soil series it ranged from 7.53 to 10.16 cmol kg^{-1} with a mean of 9.13 cmol kg^{-1} (Table 1), Barwala soil series it ranged from 10.97 to 15.38 cmol kg^{-1} with a mean of 12.85 cmol kg^{-1} and Ladwa soil series ranged from 9.39 to 15.38 cmol kg^{-1} with a mean of 11.73 cmol kg^{-1}. The overall mean range of the cation exchange capacity (CEC) varied from 8.64 to 12.85 cmol kg^{-1}. This indicates a varying mineralogical suite probably dominated by 2:1 clay minerals of the smectite or vermiculite group. The presence of mica was optically confirmed in the silt and fine sand fractions. Our findings of study are confirmed with results of Yerima *et al.* (2013).

DTPA extractable Pb, Cr, Cd, Co and Ni

The DTPA extractable lead of Balsamand soil series ranged from 0.40 to 0.98 mg kg^{-1} with a mean of 0.56 mg kg^{-1}, Isarwal soil series ranged from 0.70 to 1.38 mg kg^{-1} with a mean of 0.96 mg kg^{-1} (Table 2), Rawalwas soil series ranged from 0.44 to 1.06 mg kg^{-1} with a mean of 0.82 mg kg^{-1}. In Barwala soil series it varied from 1.08 to 2.08 mg kg^{-1} with a mean of 1.41 mg kg^{-1} and Ladwa soil series ranged from 1.04 to 1.80 mg kg^{-1} with a mean of 1.30 mg kg^{-1}. The overall mean range of the DTPA extractable lead (Pb) varied from 0.56 to 1.41 mg kg^{-1}. Guo *et al.* (2013) also found that mean concentrations of heavy metals in the top soils of Dongguan city (China) were lower than the maximum allowable concentrations (MAC). The concentrations of Cd for part of the samples exceeded the MAC and Pb concentrations in all top soils were lower than the MAC. This indicates that Pb and Cd have accumulated in agricultural soils in Dongguan city (China). The DTPA extractable chromium of Balsamand, Isarwal, Rawalwas, Barwala and Ladwa soil series, ranged from 0.01 to 0.04 mg kg^{-1} with a mean of 0.03 mg kg^{-1}, from 0.03 to 0.09 mg kg^{-1} with a mean of 0.06 mg kg^{-1}, from 0.02 to 0.08 mg kg^{-1} with a mean of 0.05 mg kg^{-1}, from 0.03 to 0.19 mg kg^{-1} with a mean of 0.12 mg kg^{-1} and from 0.09 to 0.28 mg kg^{-1} with a mean of 0.17 mg kg^{-1}, respectively. The overall mean range of the DTPA extractable chromium (Cr) varied from 0.03 to 0.17 mg kg^{-1}. Bora *et al.* (2013) also found similar results in DTPA extractable chromium of Nagoan district of Assam, the average concentration of Cd and Pb was found to be very higher and concentration

Table 2 Soluble concentrations of heavy metals content in surface layer (0-15 cm)

Soil series	DTPA extractable heavy metals (mg kg^{-1})				
	Lead	Chromium	Cadmium	Cobalt	Nickel
Balsamand	0.40-0.98 (0.56)*	0.01-0.04 (0.03)	0.01-0.20 (0.03)	0.00-0.01 (0.00)	0.02-0.06 (0.04)
Isarwal	0.70-1.38 (0.96)	0.03-0.09 (0.06)	0.01-0.02 (0.04)	0.00-0.01 (0.00)	0.00-0.04 (0.03)
Rawalwas	0.44-1.06 (0.82)	0.02-0.08 (0.05)	0.00-0.02 (0.01)	0.00-0.01 (0.01)	0.01-0.04 (0.03)
Barwala	1.08-2.08 (1.41)	0.03-0.19 (0.12)	0.00-0.02 (0.02)	0.00-0.01 (0.01)	0.01-0.54 (0.16)
Ladwa	1.04-1.80 (1.30)	0.09-0.28 (0.17)	0.00-0.02 (0.02)	0.00-0.11 (0.02)	0.02-0.19 (0.06)

*Parentheses - Mean value

of Ni and Cr was found to be much below that of the two reference values.

The overall mean range of the DTPA extractable cadmium (Cd) in different soil series varied from 0.01 to 0.04 mg kg^{-1}. The DTPA extractable cadmium of Balsamand soil series ranged from 0.01 to 0.20 mg kg^{-1} with a mean of 0.03 mg kg^{-1}, Isarwal soil series ranged from 0.01 to 0.02 mg kg^{-1} with a mean of 0.04 mg kg^{-1}, Rawalwas soil series ranged from 0.00 to 0.02 mg kg^{-1} with a mean of 0.01 mg kg^{-1}, Barwala soil series from 0.00 to 0.02 mg kg^{-1} with a mean of 0.02 mg kg^{-1} and Ladwa soil series ranged from 0.00 to 0.02 mg kg^{-1} with a mean of 0.02 mg kg^{-1} (Table 2). Yadav *et al.* (2013) and Gupta *et al.* (2008) also reported that the mean highest concentrations for Ni followed by Zn, Cu and Cd and minimum concentration of Pb was observed in soils of Naini (Prayagraj). The DTPA extractable cobalt in different soil series of study was found below 0.02 mg kg^{-1}. Guo *et al.* (2013) also found the similar results in Dongguan city (China). The overall range of the DTPA extractable nickel (Ni) varied from 0.03 to 0.16 mg kg^{-1}. The DTPA extractable nickel of Balsamand soil series ranged from 0.02 to 0.06 mg kg^{-1} with a mean of 0.04 mg kg^{-1}, Isarwal soil series ranged from 0.00 to 0.04 mg kg^{-1} with a mean of 0.03 mg kg^{-1}, Rawalwas soil series ranged from 0.01 to 0.04 mg kg^{-1} with a mean of 0.03 mg kg^{-1}, Barwala soil series ranged from 0.01 to 0.54 mg kg^{-1} with a mean of 0.16 mg kg^{-1} and in Ladwa soil series ranged from 0.02 to 0.19 mg kg^{-1} with a mean of 0.06 mg kg^{-1} (Table 2). Yadav *et al.* (2013) also found similar results in soils of Naini (Prayagraj).

Correlation matrix

To describe the relationship between different physico-chemical properties and heavy metal concentrations in soil were analyzed by Pearson's correlation coefficient (Table 3). The different quantitative variables, showed significant positive correlation between CEC and pH (0.328*), EC (0.458**), OC (0.415**), silt content (0.533**), whereas clay content with pH (0.456**), EC (0.421**), OC (0.416**), silt content (0.755**) were also found positive correlation. The lead showed positive correlation with, pH (0.355*), OC (0.388**), CEC (0.625**), silt content (0.525**) and clay content (0.612**). Between chromium and pH (0.488*), CEC (0.505**), silt content (0.558**), clay content (0.653**) also showed positive correlation. Cobalt showed positive correlation with OC (0.371**), silt content (0.368**) and clay content (0.408**). Also found positive correlation of nickel with, OC (0.289*), CEC (0.536**) and clay content (0.323*). However, negative correlation also found between sand and Pb, Cr, Co, Ni content except Cd content. Negative correlation found between clay and Cd content (0.182). Among the heavy metals correlation between Pb and Cr (0.604**), Co (0.436**) and Ni (0.430**) were positive but negative with Cd (0.137). Chromium showed positive with Co (0.289*) and negative with Cd (0.110). Cadmium showed negative correlation with Co (0.045) and Ni (0.074). Our findings are also supported by

Table 3 Pearson's correlation coefficients (r) between physico-chemical properties and heavy metal concentrations in different soil series of Hisar

Variable	pH (1:2)	EC (dS m^{-1})	OC (%)	$CaCO_3$ (%)	CEC (cmol kg^{-1})	Sand (%)	Silt (%)	Clay (%)	Pb	Cr	Cd	Co	Ni
pH (1:2)	1												
EC (dS m^{-1})	0.166	1											
OC (%)	0.004	0.245	1										
$CaCO_3$ (%)	0.282*	-0.022	-0.013	1									
CEC (cmol kg^{-1})	0.328*	0.458**	0.415**	0.013	1								
Sand (%)	-0.419**	-0.502**	-0.348*	-0.087	-0.628**	1							
Silt (%)	0.353*	0.505**	0.270	0.149	0.533**	-0.960**	1						
Clay (%)	0.456**	0.421**	0.416**	-0.018	0.676**	-0.908**	0.755**	1					
Pb (mg kg^{-1})	0.355*	0.173	0.388**	0.025	0.625**	-0.595**	0.525**	0.612**	1				
Cr (mg kg^{-1})	0.488**	0.208	0.221	-0.038	0.505**	-0.635**	0.558**	0.653**	0.604**	1			
Cd (mg kg^{-1})	0.025	-0.160	-0.206	0.277	-0.122	0.129	-0.079	-0.182	-0.137	-0.110	1		
Co (mg kg^{-1})	0.216	0.108	0.371**	0.166	0.247	-0.409**	0.368**	0.408**	0.436**	0.289*	-0.045	1	
Ni (mg kg^{-1})	0.085	0.127	0.289*	0.075	0.536**	-0.295*	0.246	0.323*	0.430**	0.206	-0.074	0.133	1

*Correlation is significant at the 0.05 level, **Correlation is significant at the 0.01 level other literatures as reported by Lv *et al.* (2015) in soils of Eastern China.

Conclusions

Based on results it may be concluded that heavy metals accumulation and their distribution in soils of alluvial and aeolian plain of Haryana are inevitable due to climatic conditions and anthropogenic activities. The mean ranges of DTPA extractable heavy metals concentration in studied samples were found below the permissible limit. Therefore, present results of the study are very useful to estimate the metal contamination and risk associated in agricultural soil to assure better soil health, crop quality and production.

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