



Research Article

COMPETITIVE ADSORPTION OF HEAVY METALS BY SOILS

BANSAL O.P.*

Department of Chemistry, D. S. College, Aligarh, 202001, Dr Bhimrao Ambedkar University, Agra, 282004, Uttar Pradesh, India

*Corresponding Author: Email - drop1955@gmail.com

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Abstract: Manner and from human activities there are huge possibilities of the change in heavy metal ratio in soil and plants during a time period. This study was conducted to evaluate the selectivity sequence and to estimate the competitive adsorption of several heavy metals (Pb, Cu, Zn, Cd and Ni) on six different soils of India in mono metal and multi metal solutions. The results denoted that the sorption isotherms of all these heavy metals on all the studied soils were characterized by Langmuir equation. Results indicated that all the soils have a maximum sorption capacity and binding strength for Pb. Metal solid/liquid distribution coefficients (K_d), which represent the adsorption affinity of the metallic cations in solution for the solid phase were also calculated, the K_d values denote that the sequence of preference was $Pb > Cu > Cd > Zn > Ni$ which may be correlated with the metal hydrolysis constant. The value of metal K_d was lower in multi metal solution than mono metal solution. The order of preference was supported by Langmuir Q_{max} values and relative sorption capacity values. The adsorption of heavy metals was significantly positively correlated with soil organic matter and soil CEC while negatively correlated with soil pH. The values of Langmuir constants, K_d and relative sorption capacity also suggest that the adsorption was in the order of soil 3 > soil 5 > soil 2 > soil 4 > soil 1 > soil 6.

Keywords: Heavy metals, Distribution coefficient, Competitive adsorption, CEC, Organic carbon

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Introduction

According to FAO [1], food security means all the people at all the times get safe nutritious sufficient food of his/her preference for an active and healthy life. Heavy metals are important environmental pollutants threatening the health of human population and native ecosystem. In agricultural soils, heavy metals, which depresses growth of plants [2], are build up mainly by irrigation of agricultural lands with urban and industrial waste water, by use of excessive fertilizers and indiscriminate use of pesticides [3,4]. Various adverse effects have been observed on soil characteristics where irrigated with waste water [5]. Under these conditions, several heavy metal cations can be available at the same time in the soils and therefore, selective retention and competitive adsorption by the soil becomes of major importance in determining their availability to plants and their mobility through the soils. Selectivity sequence of heavy metals on soils is correlated with the hydrolysis properties of the heavy metal cations [6, 7]. The capacity of soils to retain and release metals can be an important factor for prediction of environmental impact. Number of workers [8, 9] has reported that distribution coefficient (K_d) is a useful parameter for comparing the sorptive capacity and mobility for any particular ion in different soils under the same experimental conditions. As majority of published work deals with the uptake and effects of a single metal and in view of the known interactions of elements with regard to uptake and their impacts on the physiological processes of plants [10, 11]. The Present study evaluated the relative retention capacity of Pb^{2+} , Cu^{2+} , Cd^{2+} , Zn^{2+} and Ni^{2+} applied individually and in mixture to soils of Aligarh district by measuring K_d values, Langmuir constants and relative sorption capacity.

Materials and Methods

Six soil samples (S1 to S6) from cultivated land of different parts of India (0-20 cm depth) having different physico-chemical properties were selected for this study.

The soil samples were air dried at room temperature, sieved to pass through a 100 mesh sieve. Their physico-chemical properties determined by the usual soil laboratory methodology [12] are given in [Table-1]. Adsorption experiments were conducted by taking 1 g of soil samples separately in a large number of stopped conical flasks, adding various amounts (0-15 mL of 40 mg L⁻¹) of single and of cocktail solution containing nitrates of Pb, Cu, Cd, Zn and Ni in the same concentration and making up the volume to 25 mL with distilled water. The concentration 40 mg L⁻¹ was selected as there was no precipitation, and precipitation started to appear at higher concentrations. The samples were shaken for 12 h in a shaker at room temperature. Then the soil samples were centrifuged for five minutes at 3000 rpm. The supernatant liquid obtained was analysed for heavy metal concentration by atomic absorption spectrophotometer. The amount of heavy metal adsorbed was obtained from the amount added minus left in the supernatant. The distribution coefficient (K_d) was calculated as:

$K_d = \text{Equilibrium metal concentration adsorbed} / \text{Equilibrium metal concentration in solution}$

Results and discussion

The adsorption isotherms obtained for mono metal and multi metal sorption on studied soils are given in [Fig-1]. The sorption of all the studied metal ions increased with the increase of metal solution concentration and sorption from the single metal solution was much more effective than sorption of same metal from the multi metal solution, the results is in concurrent with the work of Khalfa, *et al.*, [13]. The sorption of same metal in multi metal solution was 33-50 % of single metal solution. The sorption followed the order $Pb > Cu > Cd > Zn > Ni$. The values of distribution coefficients (K_d) [Table-2], which represents the sorption affinity of the metal cations in solution for the soil solid phase, can be used to

Competitive Adsorption of Heavy Metals by Soils

Table-1 Selected Physical and Chemical properties of the soils used

Soil	Location	Organic Carbon (gkg ⁻¹)	Clay %	Sand %	Silt %	pH (1:2.5)	CEC (cmol (p+) kg ⁻¹)
S1	Bangalore	1.34	20.2	30.8	49	6.3	9.4
S2	Aligarh	1.72	13	39	48	8.9	11.2
S3	Kota	3.3	45.2	10.6	44.2	7.1	31.8
S4	Jhansi	1.52	27.2	48.2	24.6	7.6	16.5
S5	Doiawala	2.5	20.2	25.4	54.4	5.8	20.4
S6	Ludhiana	0.84	19.6	38.5	41.9	8.1	8.8

Table-2 Values of (Kd) (L g⁻¹) for sorption in six soils calculated from mono metal and multi metal sorption in solution

Metal	Concentration added (mmoles L ⁻¹)	Soil 1		Soil 2		Soil 3		Soil 4		Soil 5		Soil 6	
		Mono metal	Multi metal	Mono metal	Multi metal	Mono metal	Multi metal	Mono metal	Multi metal	Mono metal	Multi metal	Mono metal	Multi metal
Pb	0.5	47.7	26	64.3	28	100	30	53.1	27.2	75	29	44.1	25.1
	1.0	27.1	12.9	37.5	15.3	58.1	16.7	31.8	14.1	46.4	16.7	23.1	11.8
	1.5	25.7	8.5	33.6	10.4	50	12.5	28.6	9.1	40.2	11.8	20.7	7.9
	2.0	21.7	7.1	27.2	8.8	41.7	10.1	24.1	7.9	32.1	9.7	18.2	6.7
	2.5	18.4	6.7	23.1	8.1	34.4	9.9	20.3	7.4	27.1	9	15.6	6.3
	3.5	15.1	6	18.3	7.4	26.5	8.5	16.7	6.7	21.7	7.9	13	5.6
	5.0	13.3	5.2	15.9	6.1	21.3	7.1	14.6	5.6	17.7	6.6	12.2	4.9
	7.5	11.5	4.7	13.3	5.3	16.7	5.9	12.9	5	14.6	5.6	10.6	4.5
	10.0	10.3	4.1	11.8	4.6	14.7	5.2	10.9	4.3	12.7	4.9	9.7	4
	12	9.9	4	11.2	4.5	13.4	5	10.5	4.2	12	4.7	9.3	3.8
	14	8.9	3.7	10.1	4	11.8	4.7	9.4	3.8	10.9	4.2	8.5	3.4
	15	8.6	3.6	9.7	3.9	11.4	4.5	9.1	3.7	10.3	4.1	7.9	3.3
Cu	0.5	27	16	29.5	18	31.8	20	28.5	16.6	30.1	19	25	15.2
	1.0	14.1	6.4	16.7	7.9	19.6	9.7	15.3	7.1	17.8	8.8	13.9	6.2
	1.5	9.1	4.8	11.8	5.2	14.9	6.3	10.4	4.8	12.5	5.7	8.5	4.3
	2.0	7.5	3.7	9.7	4.1	12.9	4.8	8.8	3.7	10.7	4.4	6.7	3.4
	2.5	6.9	3.2	9	3.7	11.8	4.2	7.9	3.4	10.1	3.9	6.3	2.9
	3.5	6.5	2.5	7.9	3.2	10	4	7.2	2.9	8.7	3.6	5.8	2.3
	5.0	5.2	2.3	6.3	2.8	7.9	3.3	5.8	2.5	7.1	3	4.8	2.2
	7.5	4.7	2.2	5.5	2.7	6.6	3.1	5.1	2.4	5.9	2.9	4.2	2
	10.0	4.1	2.1	4.8	2.6	5.6	3	4.5	2.3	5.2	2.8	3.8	1.9
	12	4	2	4.7	2.3	5.4	2.6	4.4	2.2	5	2.5	3.7	1.8
	14	3.6	1.8	4.2	2	4.8	2.3	3.9	1.9	4.5	2.2	3.3	1.6
	15	3.5	1.7	4.1	1.9	4.7	2.2	3.8	1.8	4.4	2.1	3.2	1.5
Cd	0.5	22	13.8	24.8	17	27	19.1	23.2	15	26.2	18	21.2	13.4
	1.0	10.7	5	12.2	6.2	14.1	7.1	11.8	5.5	12.9	6.9	9.7	4.8
	1.5	7.3	3.6	7.9	4.3	9.1	4.8	7.7	3.9	8.5	4.7	6.8	3.4
	2.0	5.5	2.9	6.3	3.4	7.5	3.9	5.9	3.1	6.7	3.8	5.1	2.8
	2.5	4.5	2.6	5.6	2.9	6.9	3.3	5	2.7	6.3	3.1	4.2	2.5
	3.5	4.4	2	5.2	2.3	6.5	2.9	4.8	2.2	5.8	2.5	4	2
	5.0	3.7	1.9	4.5	2.2	5.2	2.5	4.1	2.1	4.8	2.3	3.3	1.8
	7.5	3.3	1.8	3.9	2.1	4.7	2.4	3.7	2	4.2	2.2	3	1.7
	10.0	3.2	1.7	3.7	2	4.1	2.3	3.4	1.9	3.9	2.1	2.9	1.6
	12	3.1	1.5	3.6	1.8	3.9	2.2	3.3	1.6	3.7	2	2.8	1.4
	14	2.6	1.4	3.1	1.7	3.5	1.9	2.9	1.5	3.3	1.8	2.4	1.3
	15	2.5	1.3	3	1.6	3.5	1.8	2.8	1.4	3.2	1.7	2.3	1.2
Zn	0.5	21.6	13	24	16.4	26.1	18.5	23	14.3	25	17.2	20	12
	1.0	7.9	4.1	9.7	5.5	10.7	6.9	8.8	4.8	10.1	6.3	7.1	4
	1.5	5.2	3	6.3	3.9	7.3	4.7	5.7	3.4	6.8	4.3	4.8	2.9
	2.0	4.1	2.5	4.8	3.1	5.9	3.7	4.4	2.8	5.1	3.4	3.7	2.3
	2.5	3.7	2.4	4.2	2.7	5.1	3.2	3.9	2.6	4.5	2.9	3.4	2.2
	3.5	3.2	1.8	4	2.2	5	2.5	3.6	2	4.4	2.3	2.9	1.7
	5.0	2.8	1.7	3.3	2	4.2	2.3	3	1.9	3.7	2.2	2.5	1.6
	7.5	2.6	1.6	3.1	1.9	3.9	2.2	2.9	1.8	3.3	2	2.4	1.5
	10.0	2.5	1.5	2.9	1.8	3.5	2.1	2.7	1.7	3	2	2.3	1.4
	12	2.4	1.4	2.7	1.6	3.1	2	2.5	1.5	2.9	1.8	2.2	1.3
	14	2.1	1.3	2.4	1.5	2.7	1.8	2.2	1.4	2.6	1.7	1.9	1.2
	15	2	1.2	2.3	1.4	2.6	1.7	2.2	1.3	2.5	1.6	1.8	1.1
Ni	0.5	20	8.3	22	10.3	24.2	13	21	9.1	23.2	11.7	18.1	7.3
	1.0	6.9	2.7	7.9	3.4	9.7	4.8	7.1	2.8	8.8	4.1	6.2	2.5
	1.5	4.7	2.1	5.2	2.4	6.2	3.4	4.8	2.2	5.7	2.6	4.3	1.8
	2.0	3.6	1.9	4.1	2.2	4.8	2.9	3.7	2	4.4	2.4	3.4	1.7
	2.5	3.2	1.7	3.7	2.1	4.2	2.5	3.4	1.9	3.9	2.2	2.9	1.6
	3.5	2.5	1.4	3.2	1.7	4	1.9	2.9	1.5	3.6	1.8	2.3	1.3
	5.0	2.3	1.3	2.8	1.5	3.3	1.7	2.5	1.4	3.0	1.6	2.2	1.1
	7.5	2.2	1.2	2.6	1.4	3.1	1.6	2.4	1.3	2.9	1.5	2.1	1.1
	10.0	2.1	1.1	2.5	1.3	2.9	1.5	2.3	1.2	2.7	1.4	2	1
	12	2	1	2.4	1.2	2.7	1.4	2.2	1.1	2.5	1.3	1.8	0.9
	14	1.8	0.9	2.1	1.1	2.4	1.3	1.9	1	2.3	1.2	1.6	0.8
	15	1.7	0.9	2	1	2.3	1.2	1.8	0.9	2.2	1.1	1.5	0.8

Table-3 Langmuir parameters for sorption in six soils calculated from monometal and multimetal sorption in solution

Soil	Pb				Cu				Cd				Zn				Ni			
	Monometal																			
	Qmax	b	R ²	% Sorbed	Qmax	b	R ²	% Sorbed	Qmax	b	R ²	% Sorbed	Qmax	b	R ²	% Sorbed	Qmax	b	R ²	% Sorbed
S1	42	2.1	0.958	39	31	1.3	0.974	22	28	0.85	0.962	20	25	0.76	0.948	17	23	0.88	0.964	13
S2	62	3.2	0.982	42	48	0.96	0.954	25	36	1.05	0.974	23.5	31	0.84	0.962	19.5	28	0.76	0.944	14.7
S3	80	5.4	0.964	51	62	1.8	0.936	29	51	1.35	0.944	26.5	43	0.98	0.934	23	40	1.14	0.968	17
S4	51	2.8	0.952	41	34	1.1	0.958	23.5	28	0.82	0.976	21.5	23	1.02	0.954	21	21	0.96	0.934	13.9
S5	73	4.8	0.976	46	53	1.5	0.984	27	42	1.1	0.958	25	37	0.94	0.946	18	32	1.06	0.972	15.8
S6	37	1.3	0.988	36	28	0.84	0.962	21	24	0.72	0.966	18.5	19	0.84	0.972	16	17	0.88	0.966	12.5
Multimetal																				
S1	27	1.1	0.936	21	22	0.82	0.954	12.5	20	0.78	0.942	10.5	17	0.84	0.954	9.8	15	0.72	0.938	7.4
S2	41	1.8	0.954	23.5	34	0.92	0.948	13.5	30	0.84	0.962	11.5	26	0.92	0.934	11.5	23	0.83	0.946	8.5
S3	58	3.2	0.948	26	46	1.04	0.952	16	38	1.06	0.944	13.5	33	0.98	0.952	13	29	0.88	0.964	10
S4	33	1.4	0.968	22	25	0.96	0.964	13	22	0.88	0.952	11	20	0.94	0.948	10.6	18	0.68	0.972	8
S5	47	2.5	0.952	24	35	0.88	0.934	14.5	28	0.78	0.958	12.5	24	0.74	0.936	12	20	0.82	0.954	9.1
S6	23	0.98	0.938	20	18	0.78	0.966	12	15	0.74	0.946	10	13	0.72	0.954	9	11	0.56	0.96	6.7

evaluate the sorption and retention of metallic cations in soils [14]. High values of K_d denote that that metal is more sorbed and retained by the solid, while low values of K_d denote that most metal remains in solution where it is available for transport [15]. The K_d values for mono metal and multi metal sorption were maximum for Pb and minimum for Ni. The K_d values except for Ni in multi metal solution were more than unity, showing that the studied metal ions were retained by the soils. The lowest K_d values were obtained for soil 6 and highest for soil 3 these can be related to soil organic matter, soil CEC, soil surface area and soil pH. The sorption followed the order soil 3 > soil 5 > soil 2 > soil 4 > soil 1 > soil 6. The K_d values decreased with increase in initial metal concentration. The decrease in K_d values denote the decreasing average binding energy of occupied sites as concentration increases. The sites occupied at low concentration are all of relatively high energy while sites occupied at high concentration includes both high and low energy sites. The affinity sequence of the studied metal ions is Pb > Cu > Cd > Zn > Ni. The sorption of metals depends on electrostatic forces and covalent bonding with available surface in soil [16]. The sorption of metal depends on electronegativity, ionic radius, hydrolysis constant etc. [17]. The highest affinity of all the studied soils for Pb may be due to (i) greater first hydrolysis constant (ii) higher ionic radius (iii) higher electrostatic force due to larger Misono softness value. The data of our study also showed that Cd was more adsorbed than Zn in all the soils. Similar results were also reported by Fontes, *et al.*, [18]. Nickel, despite its higher electronegativity value compared to Zn or Cd was least retained in all the studied soils. These data also denote that order of relative metal cation adsorption by the different soils was soil 3 > soil 5 > soil 2 > soil 4 > soil 1 > soil 6, showing that soil 3 has maximum ability to adsorb and store heavy metals while soil 6 has the least. The adsorption of metal cations on soil can be significantly positively correlated with soil organic matter ($r = 0.874$ to 0.958) and cation exchange capacity (CEC) ($r = 0.906$ to 0.974). There was a positive correlation between metal adsorption and surface area of soil. The adsorption of metal cations was negatively correlated with soil pH, which may be due to formation of $(M OH)^+$ at higher pH. Comparison of mono metal and competitive sorption isotherms [Fig-1] [Table-2] reveals that sorption of all the metals decreases in presence of other cations. The K_d values decreases significantly for all the metals on all the soils studied decreases 60% for Pb, 50% for Cu, 45% for Cd, 40 % for Zn and 45% for Ni. These data suggest that metals are competing for same binding sites. Data of [Fig-1] also denote that at low metal concentrations, effect of competition was not very strong. All heavy metals studied, both single and multiple initial metal solutions, were adequately described by the Langmuir equation, $C_x / m = 1 / kb + C / b$, where C is metal concentration of equilibrium solution (mg L⁻¹), b is the adsorption maxima and k is a constant related to bonding energy in soil, with $R^2 > 0.93$ as given in [Table-3]. The q_{max} , from the Langmuir equation, may be a useful parameter for comparing the potential capacity of the soil among all the studied metals, regardless of single or multiple component heavy metal

solutions Pb showed the highest value of adsorption maximum (q_{max}). On the basis on the q_{max} value, the order selectivity of these metals for the soil is $Pb^{2+} > Cu^{2+} > Cd^{2+} > Zn^{2+} > Ni^{2+}$. The selectivity order can be influenced by the valence and the ionic size of the heavy metals when hydrated. Then, smaller ions with the same valence, such as Cd compared with Pb, have higher charge densities and attract more water molecules, resulting in a larger hydrated radius [19]. Metals with higher hydrated radius exert weaker columbic forces of attraction. Therefore, Cd (0.23 nm radius) is expected to be mobile than Pb (0.187 nm radius) because of its larger hydrated radius [20]. The higher affinity of the soil for Pb may probably due to the existence of a greater number of active sites with high specificity for Pb, so, when it is present these sites would not be occupied by others cations. Moreover, the higher binding affinity coefficient b of Pb was also the greatest among all the metals studied, indicating that Pb appeared to be related to sorption at high energy surfaces with lower dissociation constants. The experimental data also confirmed the role of metal coexistence on Pb, Cu, Cd, Zn and Ni, sorption, by reducing the sorption capacity of each metal. However, the lower Q_{max} and b values may reflect both the competitive sorption among the metals and also the saturation of the sorption sites in the system due to the coexistence of other metals. To compare the sorption behaviour of metals a graph was plotted in between the relative sorption capacity (RSC) and initial metal concentration [Fig-2]. The data of [Fig-2] showed that the RSC values decreased as initial metal concentration increased the values were significantly affected by both metal ion and soil properties [21]. The RSCav (Table 3) showed that the sorption is in the order soil 3 > soil 5 > soil 2 > soil 4 > soil 1 > soil 6 denoting that sorption of metals by soils from single metal solution or multi metal solutions depends on soil organic matter, soil CEC, soil pH and mineral composition. [Fig-2] also denote that sorption of metals in mono metal solution and multi metal solutions followed the order Pb > Cu > Cd > Zn > Ni.

Conclusions

The distribution coefficient, K_d values for single solution were higher in comparison to mix-solution. K_d values in mono metal and multi metal solution was in the order Pb > Cu > Cd > Zn > Ni. These results are supported by Langmuir constants and relative sorption capacities (RSC).

Application of research: The results of this study denote that Cd, Zn and Ni may pose more threat to soils and groundwater than Pb and Cu.

Research Category: Environmental Pollution, Agricultural Chemistry

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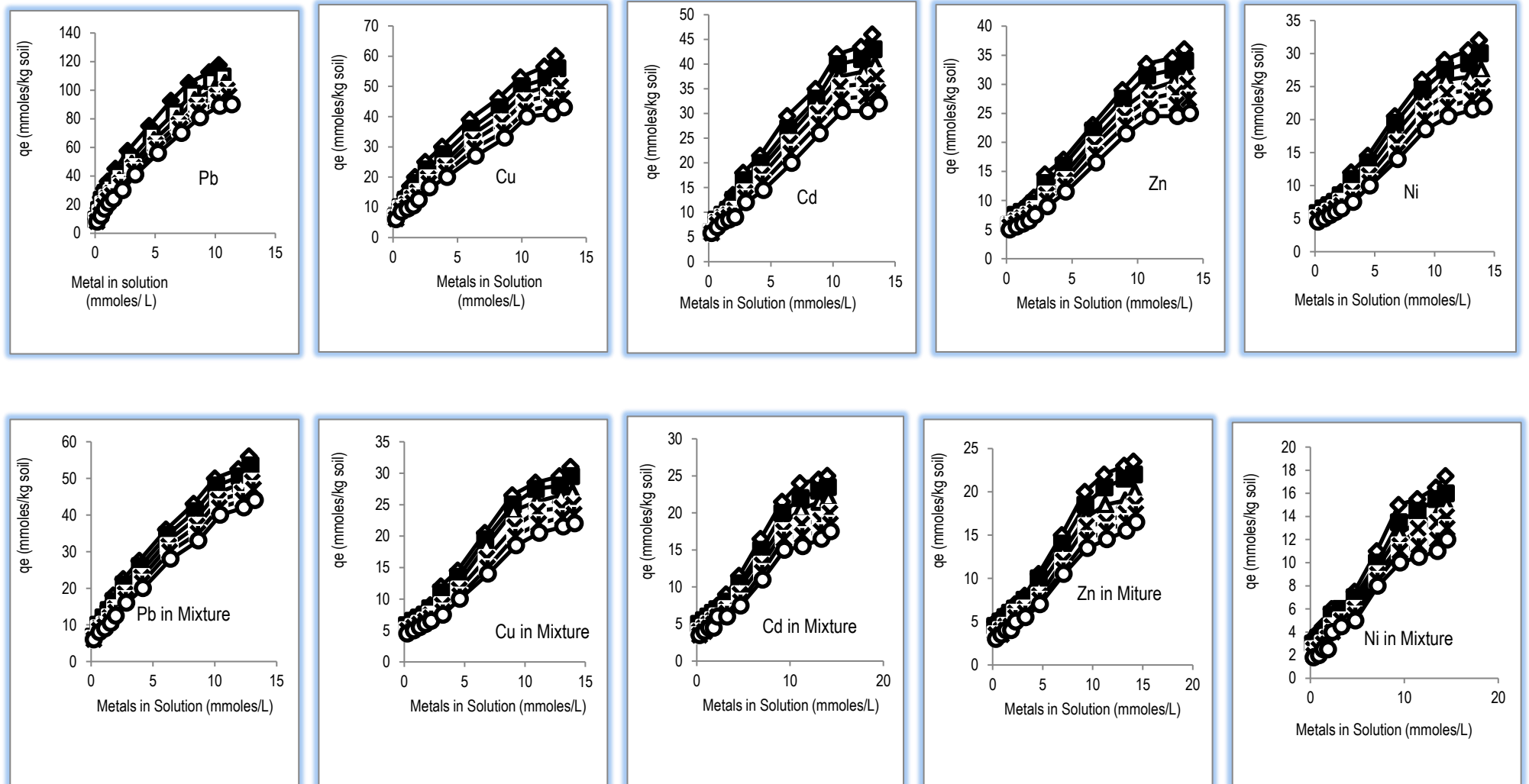


Fig-1 Monometal and competitive sorption isotherms for retention of Pb, Cu, Cd, Zn and Ni by soil 1 (*), Soil 2(△), Soil 3(◇), Soil 4 (X), Soil 5 (■) and Soil 6 (O).

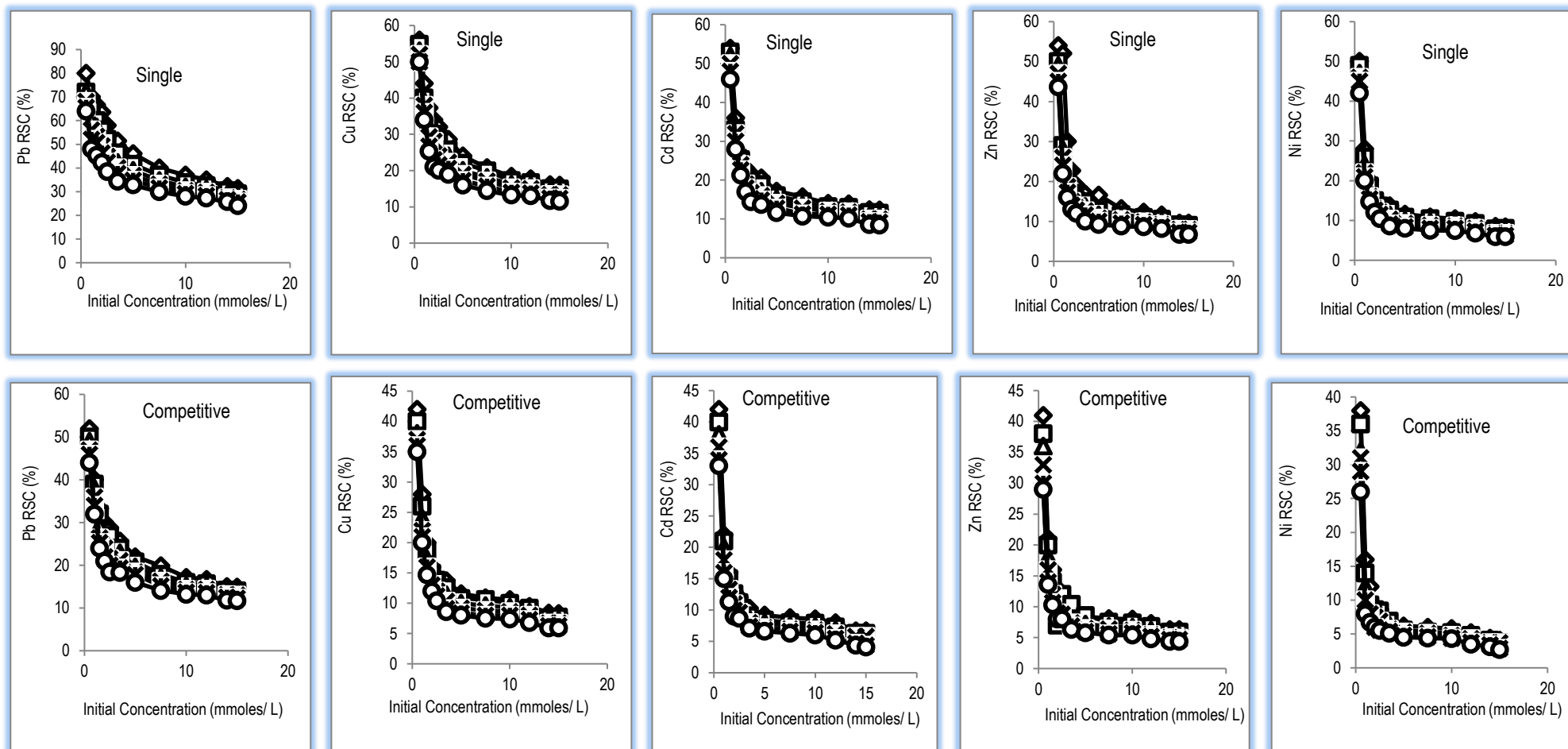


Fig-2 Relative sorption of Pb, Cu, Cd, Zn and Ni by soil 1 (*), Soil 2 (▲), Soil 3 (◇), Soil 4 (X), Soil 5 (◻) and Soil 6 (O) in single metal and multi metal solution

***Principle Investigator: O.P. BANSAL**

University: Dr Bhimrao Ambedkar University, Agra, 282004, Uttar Pradesh

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