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Sorption of Zinc and Boron in the Soils of Tamil Nadu

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Abstract: The Zn and B adsorption data clearly shows that the soils not only vary in their capacity to retain Zn and B but also in the energy with which they adsorb it. These two factors affect the ability of a soil to release Zn and B and to maintain supply in the soil solution. The rate of adsorption Zn and B study indicate that the equilibrium period of 24 h was sufficient to study the Zn and B adsorption characteristics of soils. The Elovichian model best described the kinetics of Zn and B adsorption by these soils. Though adsorption of Zn and B confirmed to both Langmuir and Freundlich adsorption isotherms, the fit was excellent with Langmuir adsorption isotherm for Zn and Freundlich adsorption isotherm for B. Clay, OM, CEC of the soils were significantly correlated with K values of Freundlich equation for B adsorption, b values of Langmuir equation for Zn adsorption. Removal of either OM or $CaCO_3$ reduced the adsorption capacity of B in the soils of Tamil Nadu. The study on the adsorption of Zn indicates that removal of OM, addition of P reduced the adsorption capacity as well as energy of its adsorption. P fertilization reduced the Zn requirement for optimum plant growth in soils of Tamil Nadu.

Key words: Zinc, Boron, Adsorption, Isotherm, Langmuir and Freundlich equations.

Introduction

The adsorption of nutrients is one of the most important solid and liquid phase interaction determining the release and fixation of applied plant nutrients. Adsorption and binding of these nutrients by soil colloidal particles are sufficiently strong to make it an important process in controlling the availability of these nutrients. As plants respond primarily to the trace elements activity in solution, understanding the mechanism of trace elements adsorption on soil materials is vital. Adsorption and the solution concentration of trace elements (Zn and B) in soil have been reported to be influenced by pH, sesquioxide, clay content and org.C. Desorption rather than adsorption is essential as it describes the actual soil reaction, which controls the mobility of trace elements, The knowledge of kinetics of adsorption reactions is essential for the effective control over the concentration of these nutrients in soil solution. Number of models have been developed to describe the kinetics of adsorption process of trace elements, but work on this line is very much limited in soils of Tamil Nadu. Zinc and B adsorption process on soil organic matter are not yet fully understood, even though many of the cultivated soils contain 1.0 to 8 g org. C kg^{-1}. The low content may have a significant effect on Zn and B adsorption and desorption in soils. P fertilization on soil rich in sesquioxide depress the Zn availability to plants (Stanton and Burger, 1970, Saeed and Fox, 1979), contrary to that P fertilization reduce zinc adsorption in soils containing low sesquioxide and organic matter.

Zinc and B adsorption varies among soil type and depends on or a combination of soil properties which correlate well with Zn and B adsorption. Simple correlation inadequately explain the relationships since correlation does not ensure direct and indirect effect relationship (Wright, 1921). Path coefficient analysis that partition into direct and indirect effects (Afifi and Clark, 1984) has been used to investigate relationships between soil properties and other measured chemical parameters in soils (Basta *et al.* 1993). Since the correlation between soil properties and adsorption parameters has not been subjected to path coefficient analysis, the path analysis has been used to describe Zn and B adsorption on soils. Literature on adsorption, kinetics of adsorption, use of path analysis to explain adsorption phenomenon suffers many limitations. Thus, need has been felt for the soils of Tamil Nadu. Hence, systematic studies have been carried out in detail on this subject with the following objectives: 1) to characterize the adsorption behaviour of Zn and B in soils with treated and untreated soils: ii) to assess the kinetics of Zn and B adsorption for providing information on the behaviour of Zn and B in soils, and iii) to investigate relationships between soil properties and other measured chemical parameters in soils by using path analysis.

Table 1. Characteristics of the soils used for Zn adsorption studies

Soil No.	Soil series	Soil class*	pH (1:2.5 soil water)	Free $CaCO_3$	Org.C	Al_2O_3	Fe_2O_3	CEC (c mol (p+) kg^{-1}	Sand	Clay	Texture**
					($g\ kg^{-1}$)				($g\ kg^{-1}$)		
1.	Kovilpatti	TC	7.8	55	3.3	47	61	52.1	455	101	c
2.	Subramaniapuram	TC	7.8	44	2.2	110	38	48.7	588	63	c
3.	Aniyur	VU	7.6	7	7.1	63	45	31.6	661	84	cl
4.	Dindigul	TP	7.8	6	3.0	119	50	63.4	331	123	c
5.	Dharmapuri	TC	7.7	19	7.2	94	46	51.7	364	181	cl
6.	Suramangalam	TC	7.9	45	5.4	64	39	38.4	657	103	cl
7.	Puvalur	TC	7.7	8	4.8	153	37	42.6	594	89	c
8.	Omandur	TU	7.8	109	4.1	157	43	38.4	732	48	scl
9.	Solampatti	EC	8.1	96	3.7	69	53	33.9	674	88	cl
10.	Govindapuram	TC	7.8	39	5.2	142	83	67.2	294	138	c
11.	Tulukkanur	TU	7.8	5	1.8	35	46	17.5	852	31	sl
12.	Palladam	TC	7.6	42	2.1	57	66	22.1	834	44	sl
13.	Dasarapatti	TC	7.5	113	4.0	82	54	58.7	246	233	c

**c = clay ; cl = clay loam; sl = sandy loam ; scl = sandy clay loam; s = sand

*EC = Entic chromustert ; TC = Typic Chromustert; TP = Typic Pellustert; TU = Typic Ustochrept, VU = Vertic Ustochrept

Table 2. Physical and chemical characteristics of soils used for B adsorption studies

Soil No.	Soil location	Classification	pH (1:2.5 soil : water)	Untreated		Treated				CEC (c mol (p+) kg^{-1}	Sand	Clay
				Free $CaCO_3$	Org.C	Free $CaCO_3$	Org.C	Al_2O_3	Fe_2O_3		($g\ kg^{-1}$)	
				($g\ kg^{-1}$)								
1	Kangeyam-palayam	Ustifluvents	7.80	16.8	5.5	3.2	0.9	40.8	49.2	23.8	671	197
2.	Kallipalayam	Paleustalfs	8.20	69.9	3.9	17.6	0.8	48.9	86.1	28.4	730	220
3.	Krishnagiri	Haplustalfs	8.00	8.4	1.8	0.4	0.5	37.9	32.1	12.8	771	132
4.	Kurumbalur	Haplustalfs	7.80	50.4	5.3	10.0	1.1	57.2	42.8	40.1	510	360
5.	Palladam	Ustochrepts	7.70	24.0	1.8	2.8	0.4	68.8	51.2	18.4	750	160
6.	Pattukkottai	Haplustalfs	6.50	4.8	5.3	1.2	0.3	138.0	113.0	35.5	709	219
7.	Pilamedu	Chromusterts	8.05	66.8	6.0	6.4	1.1	81.5	43.5	44.0	450	440
8.	Pongalivalasu	Ustochrepts	7.60	8.0	5.1	2.0	0.3	25.8	59.2	14.4	680	200
9.	Pootuthakku	Ustifluvents	7.50	12.0	5.5	1.6	0.5	78.3	41.7	34.5	720	220
10.	Somayanur	Haplustalfs	7.80	19.2	6.4	4.4	1.3	24.0	96.0	21.1	730	210
11.	Thiruvanna-malai	Ustochrepts	7.80	39.6	8.3	12.4	1.8	86.8	38.2	39.8	600	300

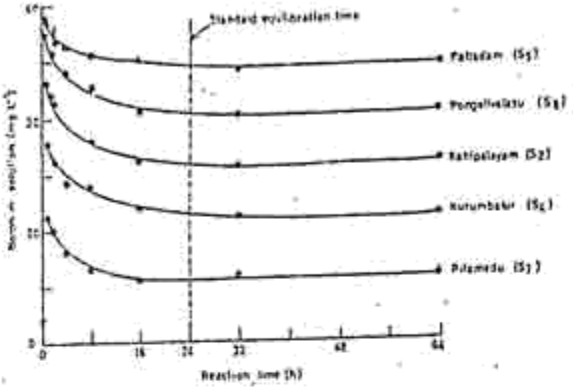

Fig.1. Boron remaining in solution in relation to reaction time

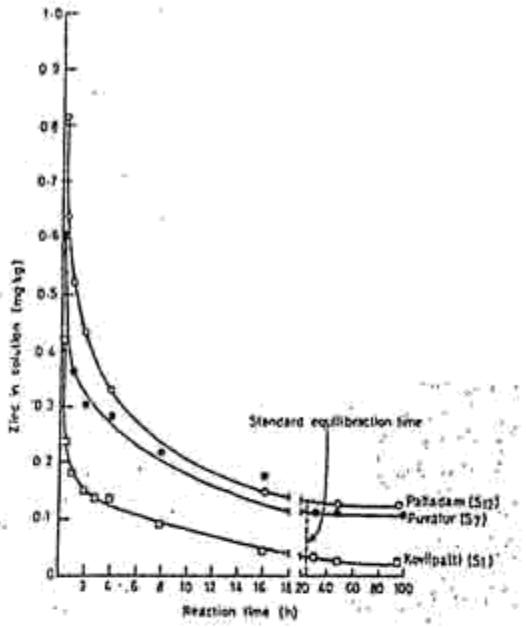

Fig.2. Zinc remaining in solution in relation to reaction time

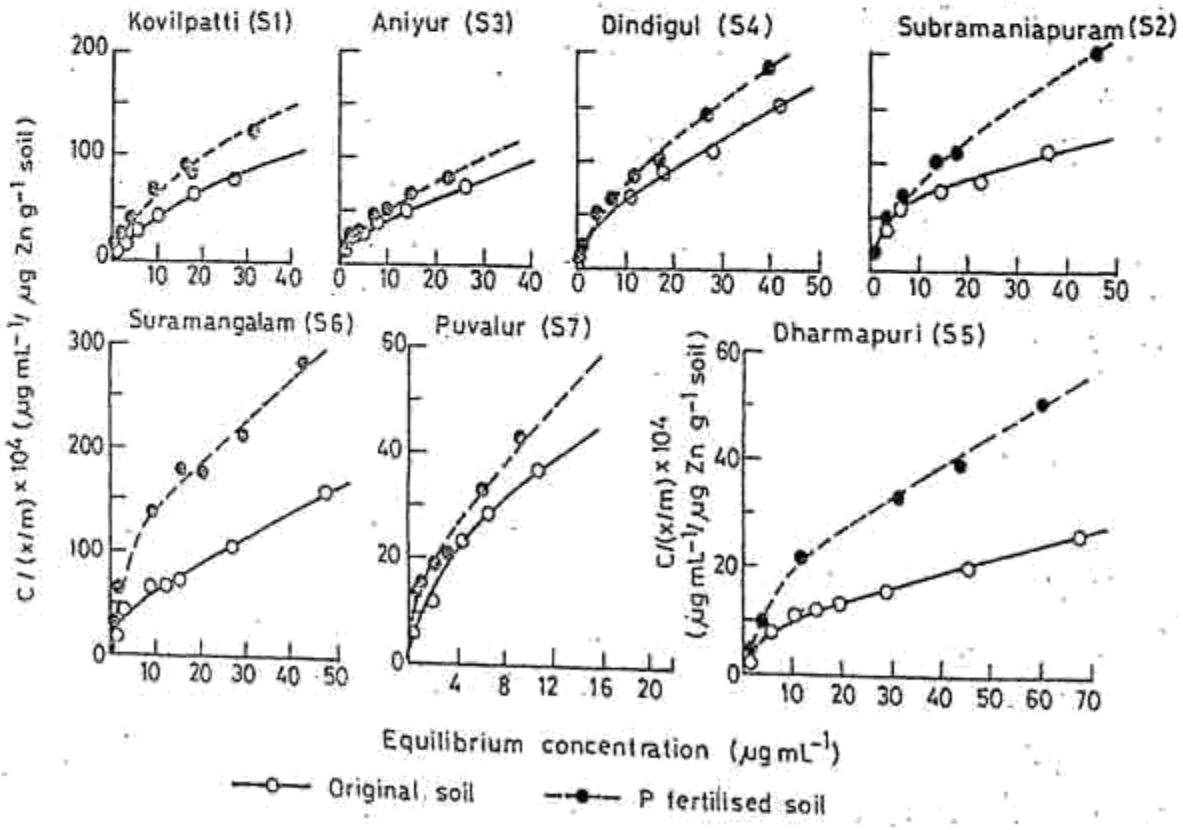

Fig.3 Langmuir isotherms for Zn adsorption in soil of Tamil Nadu

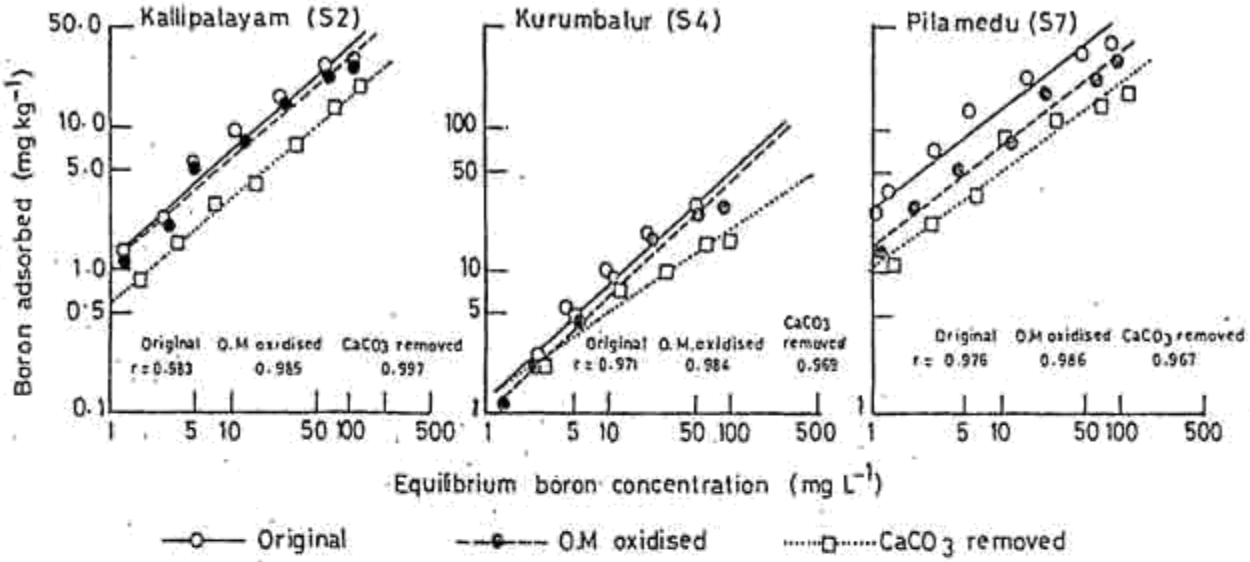

Fig.4 Freundlich isotherms for B adsorption in soils in Tamil Nadu

Materials and Methods

Soils

Adsorption experiments on Zn were conducted using thirteen surface soil samples (0-25 cm depth), representing major soil series and agroclimatic regions of Tamil Nadu and of the soil orders, Vertisol and Inceptisol and eleven surface soil samples (0-25 cm) representing four soil orders *viz.* Vertisol, Inceptisol, Alfisol and Entisol comprising the major soil series of Tamil Nadu for conducting adsorption experiments on B were collected, air dried and passed through a 2.00 mm sieve. The homogenised soil samples wee then analysed for selected physical and chemical properties (Table 1 and 2) by the methods described by Piper (1966) and Jackson (1973).

Rate of reaction

Two sets of batch studies were conducted to investigate the kinetics of the Zn and B adsorption process. In the firs set of adsorption experiments on Zn background electrolyte (20 ml, 0.01 M $CaCl_2$) containing 40 gm^{-3} Zn as $ZnSO_4.7H_2O$ was added to soil (1g) in 50 mL centrifuge tubes in triplicate. In the second set of adsorption experiments on B, calcium chloride (0.01 M) solution containing 40 mg BL^{-1} (as boric acid) was added to each of 10 g of soil in 50 mL centrifuge tubes in triplicate. The contents were shaken for 10 min and allowed to equilibrate at 27±1° for different time intervals ranging from 0.25 to 96 h for Zn and 1,2,4,8,16,32 and 64 h for boron. At the end of the reaction time, the suspension was centriftiged and filtered. The Zn concentration in the equilibrium solution of the first set of experiments was determined with Atomic absorption spectrometer and B was determined in the filtrate of the second set of experiments by the carmine method (Hatcher and Wilcox, 1950). The amount of Zn and B absorption at various time intervals were fitted to different kinetic models.

Modeling on reaction kinetics

The kinetic models used to describe Zn and B absorption from the thirteen and eight soils during 64 and 96 h respectively were tested for goodness of fit by least square regression analysis. They are :

i. Zero order : $qt=q_0 + K_0t$, where K_0 is the zero order rate constant [mg B/Zn kg^{-1} s^{-1}]

ii. First order : $Inq_t = Inq_0 + K_1t$, where K_1 is the first order rate constant (s^{-1})

iii. Second order : $1/q_t = 1/q_0 - K_2t$, where K_2 is the second order rate constant [(mg B/Zn kg^{-1})$^{-1}$.

iv. Third order : $1/qt^2 = 1/q^2j - K_3t$ where K_3 is the third order rate constant [(mg B/Zn kg^{-1})$^{-2}$ s^{-1}]

v. Parabolic diffusion : $q = \alpha Kd \sqrt{t}$ where Kd is the diffusion rate constant (mg B/Zn kg^{-1})$^{0.5}$]

Table 3. Mean and range of coefficient of determination [R^2] and standard error of estimate ISE] of various kinetic models used for Zn adsorption

Models	R^2		SE	
	Range	Mean	Range	Mean
Zero order	0.203-0.497	0.378	5.3-85.0	44.0
First order	0.196-0.459	0.355	5.3-89.6	44.1
Second order	0.188-0.417	0.310	5.3-97.7	45.7
Third order	0.181-0.375	0.311	6.0-114.3	48.8
Parabolic diffusion	0.343-0739	0.588	5.3-86.9	43.6
Elovich	0.601-0.962	0.860**	2.1-30.7	17.7

**P = 0.01

Table 4. Mean and range of coefficient of determination [R^2] and standard error of estimate [SE] of various kinetic models used for B adsorption

Models	R^2		SE	
	Range	Mean	Range	Mean
Zero order	0.387-0.581	0.464	1.13-2.59	2.05
First order	0.386-0.525	0.462	1.43-2.75	2.24
Second order	0.384-0.578	0.461	1.36-2.59	2.22
Third order	0.384-0.576	0.450	1.35-2.52	2.20
Parabolic diffusion	0.585-0.771	0.657	1.39-2.60	2.21
Elovich	0.807-0.939	0.868**	0.71-1.35	0.94

**P = 0.01

vi. Elovich : $q_t = q_0 + (1/\beta)$ In $(\alpha\beta) + (1/\alpha)$ Int, where α is the initial B/Zn adsorption rate [mg B/Zn $kg^{-1}h$] and; B/Zn description constant [(mg B/Zn $kg^{-1})^{-1}$].

In all the equations, qo and q, are the amounts of B/Zn adsorbed (mg B/Zn kg^{-1}) at time zero and t, respectively. To determine the equation that best described the adsorption of B/Zn in soils, a standard error of estimate was calculated for each equation. A relatively high value of the coefficient of determination (R^2) and low standard error was used as criteria for the best fit (Chien and Clayton, 1980). The standard error was calculated as follows:

$$SE = [\Sigma-(q-q^1)^2 / (N/2)]^{0.5}$$

Where q and ql are the measured and calculated amounts of B/Zn in soil respectively, at time t, and N is the number of measurements (Steel and Torrie, 1960).

Role of organic matter and P in Zn adsorption

Experiments were conducted in treated and untreated soil samples. The organic matter free samples were obtained by treating with hydrogen peroxide. The organic carbon was determined by wet digestion method of Walkley and Black. The soils were treated with 50 µg mL^{-1} of P as KH_2PO_4 and incubated for 30 days at 27±1°C and field moisture capacity to know the influence of P on Zn adsorption. The soils were dried, homogenized and used for adsorption studies. Available P content was estimated in treated and untreated soils (Olsen *et al.* 1954).

Role or organic matter and $CaCO_3$ in B adsorption

This was studied by investigating B adsorption in treated and untreated soil samples. Soil organic matter was removed following the procedure of Marzadori *et al.* (1991) and the content or organic carbon in soils before and after oxidation was determined by wet digestion. $CaCO_3$ was removed following the procedure of Goldberg and Forester (1991).

Adsorption studies

Zinc

The adsorption experiments were conducted in untreated and treated soils, (OM) oxidized and P fertilized soils. $CaCl_2$ solution (0.01 M; 20 mL) containing varying concentration of Zn (2.5 to 200 mg kg^{-1}) as $ZnSO_4$. $7H_2O$ was added to soil (1g). The contents were shaken and allowed to equilibrate to a predetermined equilibrium time (24 h) and centrifuged. The adsorbate concentration in the equilibrium solution was then determined as described in the case of Zn adsorption experiment. Zinc adsorption was modeled with a Langmuir adsorption isotherm.

Boron

Adsorption experiments were conducted with untreated and treated soil (OM oxidized and $CaCO_3$ removed soils) using the method described by Elrashidi and O'Connor (1982). A 10 mL solution containing varying levels of B (0 to 120 mg kg^{-1}) as boric acid in 0.01 M NaCl for $CaCO_3$ free soil and 0.01 M $CaCl_2$ for the rest of the two adsorbents was added to triplicate 50 mL centrifuge tubes containing 10 g of each soil. The contents were equilibrated using a reciprocatory shaker at 27+1°C for 24 h. At the end of the equilibrium period the tubes were centrifuged and the concentration of B in the equilibrium solution was determined. Boron adsorption was modeled with both the Langmuir and Freundlich adsorption isotherms.

Results and Discussion

Soil

The study included soils representing major soils of Tamil Nadu, varying widely in texture, free $CaCO_3$, Organic C, Al_2O_3 and cation exchange capacity (CEC). The soils possessed widely varying physio-chemical properties which provided sufficient scope for the study of Zn and B adsorption.

Kinetics of Zn and B adsorption

Zinc and B adsorption pattern were characterized in all soils by an initial quick reaction followed by a slow reaction (Fig. 1 and 2) probably due to slow diffusion of these elements in the soils. Zinc and B adsorption were almost complete after 24 h of equilibrium. The concentration of Zn and B in equilibrium solution reduced with fineness in texture. The parabolic diffusion described Zn and B adsorption over limited part of isotherm (0.25 to 4 h) for Zn, (1-8 h) for B showing discontinuity beyond 4 and 5 h reaction time respectively. This suggested that two different mechanisms are operating in respect of rate of Zn and B adsorption (Taylor *et al.* 1995). Zinc and B adsorption were best described by Elovichian kinetics (Tables 3 and 4) based on high R^2 and low SE (Chien and Clayton, 1980).

Adsorption of Zn and B

In the present study, L type adsorption isotherm curves were obtained for Zn and B adsorption in the soils of Tamil Nadu. Though the Zn and B adsorption data conformed to both Langmuir and the Freundlich equations, the fit was excellent with the Freundlich equation for B and with the Langmuir equation for Zn. Although Freundlich equation is empirical, this could be used to explain the adsorption of compounds on soils and it is best suited for heterogeneous system like the soil-nutrient- water system. While Freundlich constant K denoted the quantity of the B adsorbed when its concentration in equilibrium solution was at unity, the value of constant 'n' served as a measure of the energy of adsorption (Bailey and White, 1970).

Zinc

The adsorption maximum (b) ranged from 2.50 to 3.24 mg Zn g^{-1} soil (Table 5). There was not much variation among Inceptisols. However, the soil 13 recorded the highest 'b' value possibly due to the physical adsorption of added Zn at colloidal surfaces or $CaCO_3$, which acts as a strong adsorbent for heavy metals (Leeper, 1952). The relationship of the adsorption maxima with $CaCO_3$ ($r=0.542^*$) lends support to the above observation. Zinc adsorption was more influenced by CEC followed by clay and other soil properties. Though the use of the Langmuir isotherm is being criticized, because the exact mechanism

Table 5. Effect of oxidising soils to remove organic carbon and P added soils on Langmuir adsorption isotherm constants (b, K) for Zn adsorption in soils of Tamil Nadu

Soil No.	Original soil		OM oxidised soil		P added soil*	
	b (mg g^{-1})	K (mL μg^{-1})	b (mg g^{-1})	K (mL μg^{-1})	b (mg g^{-1})	K (mL μg^{-1})
1.	2.88	0.66	2.35	0.22	2.60	0.15
2.	3.12	0.13	2.17	0.15		
3.	3.24	0.14	2.10	0.18	2.30	0.17
4.	3.07	0.26	2.73	0.23		
5.	3.22	0.34	2.41	0.38	3.00	0.21
6.	3.10	0.74	2.81	0.48		
7.	2.52	0.22	1.66	0.13		
8.	2.66	0.30	1.79	0.22	2.40	0.18
9.	2.72	0.56	1.65	0.24		
10.	3.23	0.93	2.20	0.37		
11.	2.50	0.11	1.22	0.16	1.50	0.09
12.	3.14	0.13	1.54	0.11	2.10	0.08
13.	3.05	0.73	2.69	0.35	2.90	0.25

*Seven soils were used for P treatment

Table 6. Effect of oxidising soils to remove organic carbon and treatment to remove $CaCO_3$ on Freundlich adsorption isotherm constants (K, n) for B adsorption in soils of Tamil Nadu.

Soil No.	Original soil		OM oxidised soil		$CaCO_3$ removed soil	
	K (mg g^{-1})	n	K (mg g^{-1})	n	K (mg g^{-1})	n
1	1.04	1.46	0.61	1.39	0.43	1.24
2.	1.20	1.33	1.13	1.37	0.55	1.32
3.	0.18	1.38	0.14	1.28	0.16	1.36
4.	1.28	1.26	1.10	1.27	1.22	1.62
5.	0.38	1.59	0.38	1.66	0.30	1.71
6.	1.07	1.30	0.46	1.24	1.02	1.55
7.	1.96	2.30	1.64	1.47	1.18	1.56
8.	1.19	1.31	0.56	1.64	0.66	1.69
9.	1.30	1.32	0.55	1.29	1.14	1.80
10.	1.09	1.33	1.06	1.39	0.75	1.22
11.	1.43	1.14	0.35	1.09	0.70	1.31

of adsorption is not known, the adsorption maxima may be used with confidence to relate the soil properties (Harter and Baker, 1977; Shuman, 1988; Krishnasamy *et al.* 1997). The soil properties (clay, silt, $CaCO_3$, Org.C and CEC) except Org.C significantly correlated with adsorption maxima.

Adsorption of Zn on the organic matter oxidized soils showed a considerable reduction as compared to the corresponding original soils (Fig3; Table 5). This might be due to the decrease in adsorption capacity of soils by the oxidation of organic matter. Reduction in Zn adsorption from the original soils was not proportionate to the quantity of organic matter oxidized. The difference in the quality of organic matter would be the reasons for such variations (Prakhorov and Gromova, 1971). The soils of Pilamedu, Suramangalam, Dindigul and Dasrapatti did not show the expected reduction in Zn adsorption and this might be due to the continued influences of clay and $CaCO_3$ contents of the soils, which over ride the effect of organic matter on Zn adsorption.

Phosphatic fertilization reduced Zn adsorption in all soils. The percentage of reduction of Zn adsorption capacity due to P fertilization was 6.5 to 40.8 (Table 5). The results indicated that low clay, $CaCO_3$ and R_2O_3 contents affect the magnitude of P influence on Zn adsorption in soil 5, 6 and 2. The effect of P was more pronounced in the soil with low $CaCO_3$ content (Saeed, 1977). Hence this suggests that P induced Zn deficiency could not be ascribed to precipitation of Zn as insoluble Zn-P compounds in soils. The increased Zn solubility with P fertilization is the evidence that P-Zn interaction does not reside in the growing medium external to plant roots.

Boron

The quantities of B adsorbed when plotted as a function of equilibrium concentration on log-log scale resulted in a straight line (Fig.4). Boron adsorption was successfully described with the Freundlich model. Similar success with the Freundich model has been reported by many workers (Elrashidi and O' Connor, 1982; Nicholaichuk et al. 1988). Freundlich 'K' values for the adsorption reaction of eleven soils ranged from 0. 18 to 1.96 mg B kg^{-1} (Table 7). The values of K obtained are in consonance with the results obtained by Elrashidi and O' Connor (1982) and Nicholaichuk *et al.* (1988). This may be attributed to the high CEC, clay and sesquioxide content of all the soils studied which play a key role in B adsorption (Goldberg and Glaubig, 1985). Among the soils studied, soils 3 and 5 recorded low K values, as they were sandy in texture and low in CEC. Maximum adsorption capacity appeared to increase with the fineness of the soil texture (Biggar and Fireman, 1960). The clay and organic carbon content in the soil were significantly related with the K values of Freundlich equation ($r = 0.839^{**}$ and $r = 0.787^{**}$, respectively). Similar results were expressed by Elrashidi and O' Connor (1982). Several investigators (eg. Yermiyahu *et al.* 1995) have indicated that organic matter is one of the active factors in B adsorption.

The K values of Freundlich adsorption isotherms were generally lower in soils treated to remove organic matter than that of original soils. This showed that organic adsorptive surface had greater affinity to retain B than mineral surfaces. Though there was a reduction in the adsorptive capacity (K values) due to oxidation of organic matter, the trend of K values followed the same pattern as that of original soils. The formation of complexes between dihydroxy organic compounds and B can presumably explain the observed behaviour, showing that organic carbon plays an important role in B adsorption. The results were also consistent with those of Elrashidi and O' Connor (1982). Removal of $CaCO_3$, led to the marked reduction in adsorption capacity (K values) for all the soils. Similar observations were recorded by Goldberg and Forster (1991). Although B adsorption decreased with $CaCO_3$ removal no significant correlation was observed between $CaCO_3$ and K values of Freundlich adsorption isotherm in the present study. This was probably a result of limited $CaCO_3$ value range within the soils studied.

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