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Removal of hexavalent chromium Cr (VI) by adsorption in blended lateritic soil

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Abstract. Hexavalent chromium [Cr (VI)] adsorption on lateritic soil and lateritic soil blended with black cotton (BC) soil, marine clay and bentonite clay were studied in the laboratory using batch adsorption techniques. In the present investigation the natural laterite soil was blended with 10%, 20% and 30% BC soil, marine clay and bentonite clay separately. The interactions on test soils have been studied with respect to the linear, Freundlich and Langmuir isotherms. The linear isotherm parameter, Freundlich and Langmuir isotherm parameters were determined from the from the batch adsorption tests. The adsorption of Cr (VI) on natural laterite soil and blended laterite soil was determined using double beam spectrophotometer. The distribution coefficients obtained were 1.251, 1.359 and 2.622 L/kg for lateritic soil blended with 10%, 20% and 30% BC soil; 5.396, 12.973 and 48.641 L/kg for lateritic soil blended with marine clay and 5.093, 8.148 and 12.179 L/kg for lateritic soil blended with bentonite clay respectively. The experimental data fitted well to the Langmuir model as observed from the higher value of correlation coefficient. Soil pH and iron content in soil(s) has greater influence on Cr (VI) adsorption. From the study it is concluded that laterite soil can be blended with clayey soils for removing Cr (VI) by adsorption.

Keywords: blended lateritic soil; hexavalent chromium; batch tests; adsorption isotherms

1. Introduction

Chromium commonly enters the environment from the effluents of various industries such as metallurgical, refractory, chemical pigments, electroplating, tanning and many others (Diatta and Kocialkowski 1997). It will cause environmental pollution when it is released into soil and water. Among the two oxidation states of chromium [trivalent, Cr (III)] and hexavalent chromium, [Cr (VI)] hexavalent chromium is very toxic even at concentration as low as 0.05 mg/L in drinking water (Das *et al.* 2013). Cr (VI) is highly mobile and poses a great risk of ground water pollution (Babu and Gupta 2001). The risk of environmental pollution and also health impact due to the carcinogenic nature of Cr (VI) several treatment methods have been developed and implemented for its treatment purpose. Adsorption is one of the most popular treatment methods and this method is also applicable in landfill liners to reduce the mobility of contaminant to the groundwater (Das *et al.* 2013). Adsorption is a mass transfer operation in which substances present

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in a liquid phase are adsorbed or accumulated on a solid phase and thus removed from the liquid (Tembhurkar and Dongre 2006). Soils containing high clay content are most commonly used for the adsorption of heavy metals due to their large surface area, high cation exchange capacity (Dube *et al*. 2001) and strong adsorption capacity (Kumar and Mukherjee 2013). Lateritic soil has been proved to be an effective adsorbent for the removal of various contaminants like arsenic (Maji *et al*. 2007), phosphorous (Patil Mansing and Raut 2013), sulphate (Gosh and Dash 2012), fluoride (Osei *et al*. 2015) etc. The adsorption of contaminants in the soils are influenced by physicochemical properties of the soil such as amount of clay and organic fraction (Diatta and Kocialkowski 1997, Dube *et al*. 2001), pH (Dube *et al*. 2001, Patil Mansing and Raut 2013), cation exchange capacity (Kumar and Mukherjee 2013) and also chemical composition of soil (Dube *et al*. 2001).

The application of adsorption in the removal of Cr (VI) have been studied by many researchers using various adsorbents like clayey soils (Agnieszka *et al*. 2012, Wanees *et al*. 2012, Ajouyed *et al*. 2011), activated carbon from coconut shell (Veena Devi *et al*. 2012), rice husk ash (Naeem *et al*. 2009), activated neem leaves (Babu and Gupta 2001) etc. However, not much study has been done on blended lateritic soil to evaluate its potential to adsorb Cr (VI). Lateritic soil can be blended with other natural materials to achieve the desired surface properties for best immobilization of contaminants and to act as a suitable landfill liner. Bennour (2012) reported that when bentonite is hydrated with water it exhibits large surface area. Hydration causes separation of microscopic platelets in its into a porous structure containing positive and negative charges. This ionic charge and porous structure helps bentonite to adsorb contaminants. Bentonite can be expected to fill all the voids in soil due to high swelling capacity under fully saturated condition, thereby reducing the hydraulic conductivity. Due to low hydraulic conductivity they suggested that by using soil-bentonite mixture as a landfill liner, thickness of liner can be reduced effectively. BC soil with additives like lime, cement and fly ash can be used as a liner material due to high cation exchange capacity and better sorptive ability (Mohammed and Naik 2011). Marine clay is microcrystalline in nature and it is composed of clay minerals like chlorite, kaolinite and illite and non-clay minerals like quartz and feldspar (Koteswara Rao *et al*. 2012). Marine clay can effectively retain heavy metals like zinc, lead, iron, cadmium and copper. The adsorption capacity of marine clay varies with the metal composition in it (Maji *et al*. 2007).

The purpose of this study is to determine the adsorption characteristics of Cr (VI) on lateritic soil blended with commercially available bentonite clay, BC soil and marine clay by batch adsorption techniques. Both linear and non linear adsorption isotherms have been developed from the experimental results to compare the adsorption coefficients of lateritic soil blended with these soils. This study will help in developing remediation materials for removing Cr (VI) in effluents as well as for the immobilization of Cr (VI) through the liner into the ground water.

2. Materials and methods

Malaysia, being one of the fast developing countries has enjoyed remarkable growth over the last few decades, with industrialization, agriculture and tourism playing leading roles in this success story.

2.1 Materials

The laterite soil used in the present study was collected from the adjacent areas of National

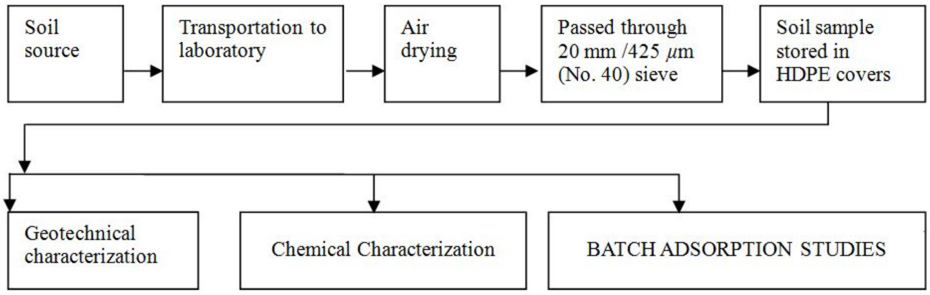


Fig. 1 Schematic diagram of various tests conducted on soils

Institute of Technology Karnataka (NITK) campus, Surathkal (Latitude 13.0092°N and Longitude 74.7937°E), BC soil was collected from North Karnataka (Latitude 15° 21' 0" N and Longitude 75° 10' 0" E), marine clay from Mangalore (Latitude 12° 51' 50" N and Longitude 74° 50' 7" E) and commercially available bentonite was procured from Bangalore. All the soil samples were collected from open excavations as per IS: 2720 (Part 29) – 1975 (Reaffirmed 1988). Depth of sampling was about 0.5 m to 1 m from natural ground level. After sampling the soil samples were air dried in shade (for about 24-36 hours). The air dried soil samples, passed through 425µm (No. 40) sieve were subject to further analysis. The flow chart in Fig. 1 shows the schematic diagram of various tests conducted on test soils.

For the present study, a stock solution of Cr (VI) was prepared synthetically for investigating adsorption characteristics of blended lateritic soil. About 2.828 g potassium dichromate ($K_2Cr_2O_7$) was dissolved in 1L distilled water to get a stock solution of 1000 mg/L. From this an intermediate solution of concentration 100 mg/L was prepared. The desired low and high concentrations of chromium solution were prepared by diluting the intermediate and stock solution respectively. After adsorption, analysis of hexavalent chromium in solution was done by colorimetric method using double beam spectrophotometer AU-2701. Detection wavelength used is 540 nm.

2.2 Batch adsorption tests

Batch adsorption tests were carried out in accordance with USEPA (1987) to evaluate the adsorption capacity of lateritic soil blended with BC soil, marine clay and bentonite clay. The experiments were conducted by mixing air dried soil samples and aqueous solution of $K_2Cr_2O_7$ for various initial concentrations ranging from 0.5 – 100 mg/L. In the present investigation the natural lateritic soil was blended with 10%, 20% and 30% BC soil, marine clay and bentonite separately. The mixture was agitated at 120 rpm in a rotary shaker for a period of 8 hours. After attaining equilibrium concentration the solution was filtered through whatman No. 2 filter paper using vacuum filter pump. The pH of the solution was noted before and after adsorption. The uptake of Cr (VI) by laterite soil particles was calculated using Eq. (1).

$$q = \frac{(C_i - C_e)V}{m} \tag{1}$$

where q, the amount of adsorbate adsorbed (mg adsorbate/kg of adsorbent); C_i and C_e are the initial and final concentration of adsorbate (mg/L); V, volume of solution taken (L); and m, mass of adsorbent taken (kg). Finally, the obtained batch adsorption test results were used to develop adsorption isotherms (*v.i.z* linear and non linear). The adsorption coefficients obtained were then compared so as to know which isotherm fits well the obtained experimental data.

3. Results and discussions

3.1 Properties of test soils

The physico-chemical properties of test soils are shown in Table 1. The pH results of lateritic soil and marine clay indicate that these soils are acidic in nature (pH 5.49 and 3.46) and that of BC soil was basic (pH 8.3) while the pH of bentonite clay is 6.86. The number of positively charged exchange sites increases with decreasing soil pH and because of the anionic nature of Cr (VI), its adsorption on soil surface increases with increase in cation exchange sites. The iron and alumina content in soil also enhance the adsorption of Cr (VI), since cations in soil favour adsorption of Cr (VI). In the soils tested the liquid limit of bentonite was found to be higher due to the large amount of clay content in it. Since the plasticity index of lateritic soil is less than 10% it should be blended with clayey soils in suitable proportions for using as a liner material in landfill.

3.2 Adsorption isotherms

Adsorption of Cr (VI) on lateritic soil blended with BC soil, marine clay and bentonite clay were analyzed using linear and non-linear adsorption isotherms. Langmuir and Freundlich isotherms provide information on the adsorption capacity of adsorbent. The isotherm models were applied to experimental data to establish the relationship between contaminant concentration and

Table 1 Physico- chemical properties of test soils

Properties	Soils			
	Laterite	Black cotton	Marine clay	Bentonite
Specific gravity (G_s)	2.65	2.45	2.46	2.53
Liquid limit (%)	31	63	40	200
Plastic limit (%)	24	30	25	44
Plasticity index (%)	7	33	15	156
Sand (%)	51	32	17	3
Silt (%)	23	44	76	43
Clay (%)	2	24	7	54
pH	5.49	8.37	3.46	6.86
Conductivity (μS/cm)	245	923	3.52	1.75
Organic matter (%)	0.86	0.54	0.69	0.024
Loss on ignition (%)	17.42	10.85	13.48	14.53
CEC (meq/100 g)	10	32	15	40
SiO_2 (%)	48.68	51.44	21.68	45.94
Al_2O_3 (%)	0.99	9.39	8.25	8.03
Fe_2O_3 (%)	37.92	27.57	58.9	27.65

Table 2 Langmuir adsorption parameters

Soil	Low concentration range (0.5-10 mg/L)			High concentration range (0.5-100 mg/L)		
	α	β (mg/kg)	R^2	α	β (mg/kg)	R^2
Lateritic soil	0.19	39.52	0.99	0.12	62.50	0.99
Lateritic soil + 10% BC soil	0.34	11.54	0.95	0.15	23.09	0.95
Lateritic soil + 20% BC soil	0.48	12.82	0.96	0.24	22.67	0.95
Lateritic soil + 30% BC soil	0.21	25.97	0.99	0.13	40.16	0.99
Lateritic soil + 10% marine clay	0.50	34.60	0.99	0.45	35.21	0.99
Lateritic soil + 20% marine clay	1.92	40.00	0.99	2.14	66.67	0.98
Lateritic soil + 30% marine clay	1.90	66.67	0.99	1.00	125.00	0.98
Lateritic soil + 10% bentonite	0.48	43.37	0.97	0.22	83.33	0.95
Lateritic soil + 20% bentonite	0.57	47.61	0.93	0.29	83.33	0.93
Lateritic soil + 30% bentonite	0.88	71.43	0.94	0.39	142.85	0.93

α - adsorption constant related to the binding energy; β - maximum amount of solute that can be adsorbed

adsorption. Langmuir isotherm model fitted well with the obtained experimental data and is discussed in the following sections. Experimental data pertaining to the linear and Freundlich isotherms are presented in *Appendix A*.

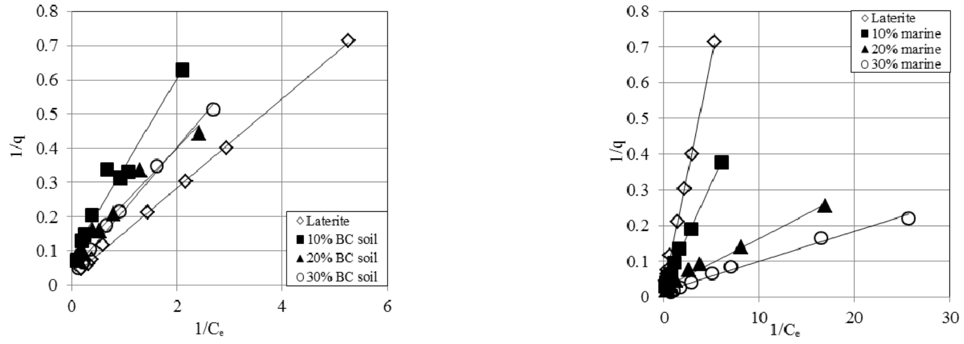
3.2.1 Langmuir adsorption isotherms

Eq. (2) shows the linear form of Langmuir adsorption isotherm. The low concentration test data of linearized Freundlich isotherm are plotted in Figs. 2(a)-(c) and the high concentration test results (all test data) in Figs. 3(a)-(c) respectively.

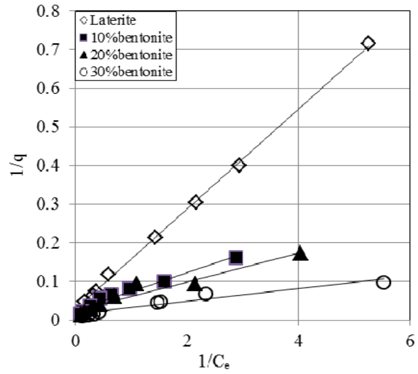
$$\frac{1}{q_e} = \frac{1}{\beta} + \frac{1}{\alpha \beta C_e} \tag{2}$$

The corresponding values of β and α for each type of soil are shown in Table 2.

The β value indicates the adsorption capacity of Cr (VI) which ranges from 11.54 mg/kg to 25.97 mg/kg for laterite-BC soil blend, 34.60 mg/kg to 66.67 mg/kg for laterite-marine clay blend and 43.37 mg/kg to 71.43 mg/kg for laterite-bentonite blend. It indicates that lateritic soil blended with 30% bentonite has greater adsorption capacity than lateritic soil blended with marine clay and black cotton soil for low concentrations of Cr (VI) solutions. It may be due to the large surface area provided by bentonite and greater availability of adsorption sites. The porous structure and clay minerals helps the bentonite to adsorb more contaminants. Also at low pH, there is an electrostatic attraction between metal ions in soil surface and anionic form of Cr (VI). The R^2 values in Table 2 indicate that the test results are most fitted to Langmuir model for all test soils. It reveals the monolayer adsorption of Cr (VI) on these blends of lateritic soil. Lateritic soil blended with bentonite shows higher adsorption capacity at high concentration of Cr (VI) (see Table 2). Also, observed from Table 2 that lateritic soil blended with BC soil and marine clay show some adsorption of C(VI) solution.

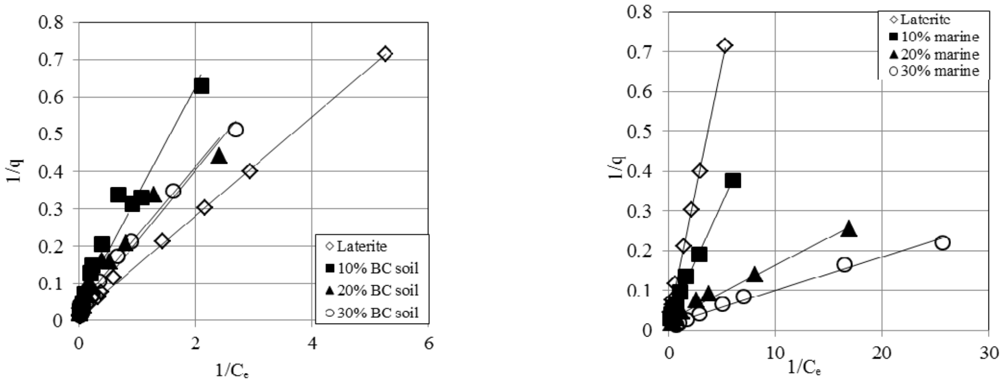


(a) Laterite blended with BC soil (b) Laterite soil blended with marine clay



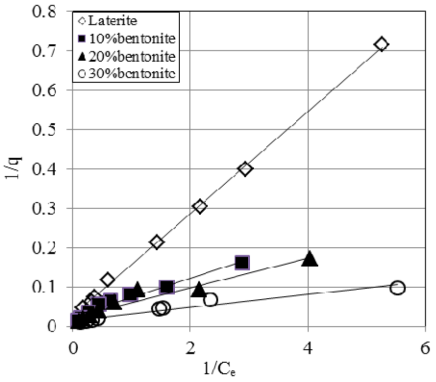
(c) Laterite soil blended with bentonite clay

Fig. 2 Langmuir adsorption isotherm for blended soil-Cr (VI) interaction



(a) Laterite blended with BC soil (b) Laterite soil blended with marine clay

Fig. 3 Linearized Langmuir adsorption isotherm for blended laterite soil-Cr (VI) all test data



(c) Laterite soil blended with bentonite clay

Fig. 3 Continued

4. Conclusions

In this study the experimental data of batch adsorption tests were fitted to linear, Freundlich and Langmuir models to determine the extent of hexavalent chromium adsorption on laterite soil blended with 10%, 20% and 30% BC soil, marine clay and bentonite clay respectively.

- The study revealed that laterite soil blended with clayey soils can be used for removing Cr (VI) by adsorption.
- The order of the adsorption of Cr (VI) on blended test soils were bentonite caly > marine caly > BC soil.
- Lateritic soil blended with marine clay has high distribution coefficient (k_d). The distribution coefficients of lateritic soil blended with marine clay vary in the range 5.39 to 48.64 L/kg. Low pH and cation content especially iron and alumina in soil favors the removal of Cr (VI) from aqueous solution.
- A significant finding from the study is the observation that the various adsorption parameters obtained for blended laterite soil showed that Langmuir isotherm model fitted well with obtained experimental data, based on the high values of the coefficient of correlation R^2 obtained from the linear plots.
- Low concentration results may be used to estimate the adsorption capacity of Cr (VI) in test soils.

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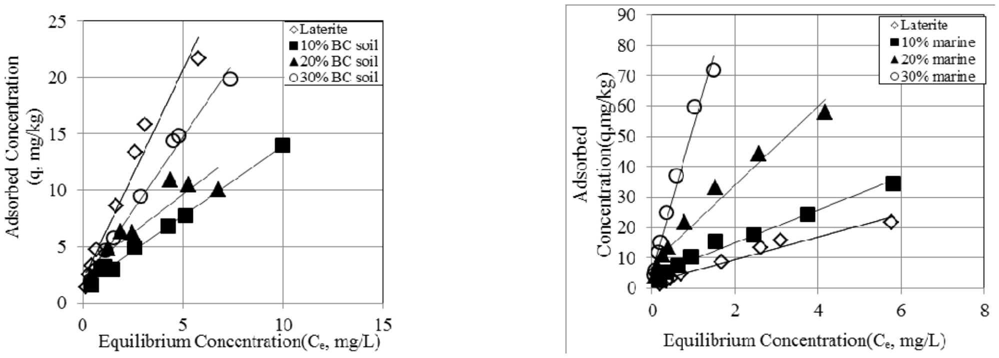
Appendix A

3.2.1.1 Linear adsorption isotherm

Linear adsorption isotherm is mostly valid only at low concentration range (i.e., 0.5-10 mg/L) of Cr (VI) soil interaction. Fig. 4(a) shows adsorption test results of lateritic soil blended with BC soil. Similarly Figs. 4(b) and (c) shows adsorption test results of lateritic soil blended with marine clay and bentonite clay respectively. In all these cases the linear adsorption isotherm is established by plotting a graph of q vs C_e (Figs. 4(b)-(c)). The relationship can be expressed as

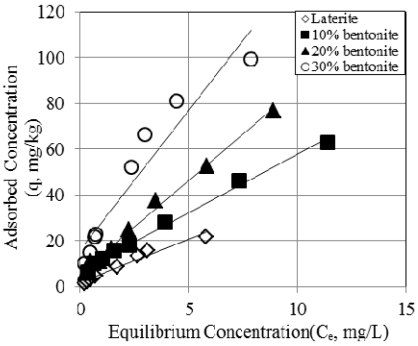
$$q = k_d C_e \tag{3}$$

From Figs. 4(b)-(c) it is observed that at low concentrations the adsorption test results of blended lateritie soil can be approximated by a linear adsorption isotherm. The slope of the line is the partition coefficient k_d. The corresponding values of k_d for lateritic soil blended with BC soil, marine clay and bentonite clay are presented in Table 2A. As observed from the experimental results (presented in Table 2A), the value of distribution coefficient (k_d) increases as the percentage of blend increases. However, the k_d value of laterite- BC soil blend is lesser compared to that of lateritic soil. It may be due to the basic nature of BC soil. The presence of OH$^-$ in soil



(a) Laterite blended with BC soil

(b) Laterite soil blended with marine clay



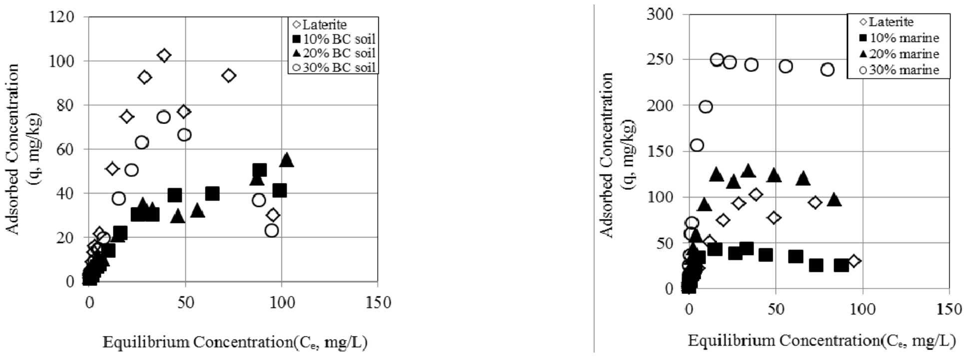
(c) Laterite soil blended with bentonite clay

Fig. 4 Linear adsorption isotherm

reduces the anion adsorption due to the hindrance of negative charges. It was found that k_d value is much higher for laterite-marine clay blend (48.64 L/kg) than laterite-bentonite blend (12.17 L/kg). It indicated the greater affinity of Cr (VI) on laterite-marine clay blend. It may be due to the high iron content and acidic nature of marine clay (see Table 1). These both properties enhance the affinity of Cr (VI) on soil due to the anionic nature of Cr (VI). Hexavalent chromium is strongly adsorbed in the presence of iron and alumina favorably in acidic and neutral media.

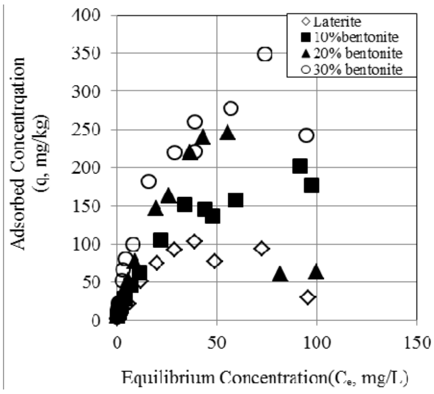
All the test data for linear adsorption isotherm are shown in Figs. 5(a)-(c). From Figs. 5(a)-(c) (for lateritic soil blended with BC soil, marine clay and bentonite clay) it is observed that at low concentrations sorption increases with equilibrium concentration until it reaches a certain threshold value. The decrease in adsorption at high concentration is possibly due to the lack of adsorption sites for an increased number of Cr (VI) ions for the fixed amount of adsorbent. Therefore the threshold value for blended laterite soil was achieved at low concentrations. Thus, the low concentration results may be used to estimate the adsorption capacity of Cr (VI) in test soils. Also, lateritic soil blended with 10% marine clay show slightly lesser adsorption. So, for obtaining better adsorption results of Cr (VI) lateritic soil should be blended with more than 10% of marine clay.

The experimental results of batch adsorption tests (linear and Freundlich isotherms) are shown in Tables 2A and 3 respectively for low concentration and high concentration Cr (VI) solutions.



(a) Laterite blended with BC soil

(b) Laterite soil blended with marine clay



(c) Laterite soil blended with bentonite clay

Fig. 5 Linear adsorption isotherms for all test data

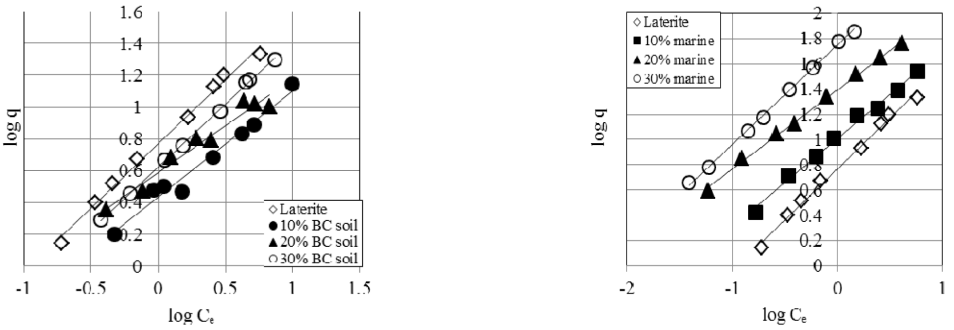
3.2.1.2 Freundlich adsorption isotherm

The linearized form of Freundlich isotherm is shown in Eq. (4). The low concentration test data of linearized Freundlich isotherm are plotted in Figs. 6(a)-(c) and the high concentration test results or all test data in Figs. 7(a)-(c).

$$\log q = \log k_F + \frac{1}{n}\log C_e \tag{4}$$

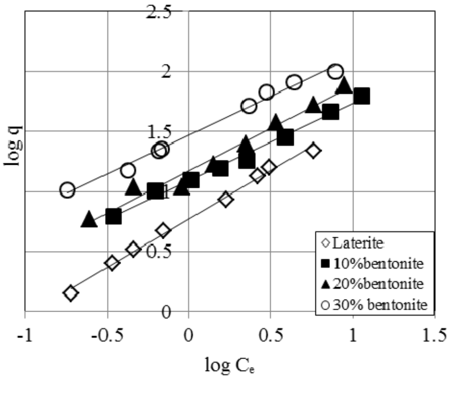
The constant k_F is an approximate indicator of adsorption capacity and $1/n$ is a function of the intensity of adsorption in the adsorption process. Tables 2A and 3 shows that '*n*' values (for low concentration and high concentration test results) are greater than one for lateratic soil blended with BC soil, marine clay and bentonite clay indicate the favorable adsorption of Cr (VI) on test soils. The adsorption parameter k_F is higher for laterite blended with 30% marine clay (40.36 mg/kg) for al test data. The variation in adsorption capacity is may be due to the difference in soil properties.

In Figs. 7(a)-(c) Freundlich isotherms of high range concentrations of Cr (VI) show that the amount of Cr (VI) adsorbed decreases with increase in concentration of contaminant. Since, energically less favorable sites in soils become involved with increasing concentration of Cr (VI)



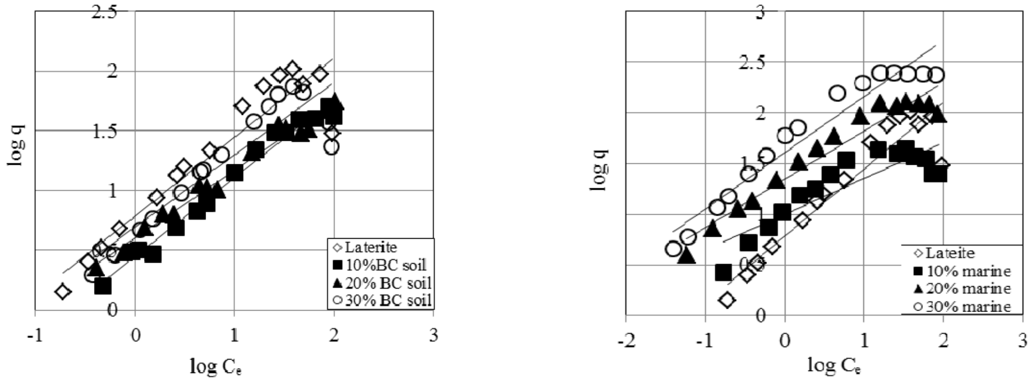
(a) Laterite blended with BC soil

(b) Laterite soil blended with marine clay



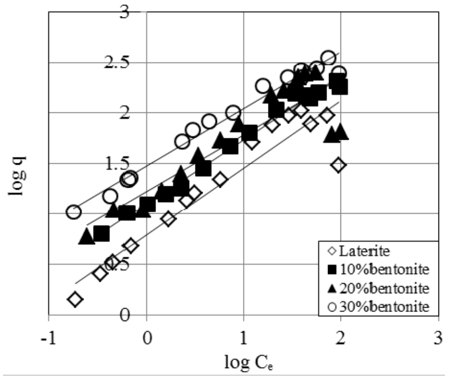
(c) Laterite soil blended with bentonite clay

Fig. 6 Freundlich adsorption isotherm for blended soil-Cr (VI) interaction



(a) Laterite blended with BC soil

(b) Laterite soil blended with marine clay



(c) Laterite soil blended with bentonite clay

Fig. 7 Linearized Freundlich adsorption isotherm for blended laterite soil-Cr (VI) all test data

Table 2A Adsorption parameters for low concentration range (0.5-10 mg/L)

Soil	Linear isotherm		Freundlich isotherm		
Lateritic soil	k_d (L/kg)	R^2	k_F (mg/kg)	n (kg/L)	R^2
Lateritic soil + 10% BC soil	3.76	0.96	6.39	1.29	0.99
Lateritic soil + 20% BC soil	1.25	0.99	2.71	1.49	0.99
Lateritic soil + 30% BC soil	1.36	0.85	3.86	1.69	0.96
Lateritic soil + 10% marine clay	2.62	0.98	4.21	1.26	0.97
Lateritic soil + 20% marine clay	5.40	0.97	10.20	1.43	0.99
Lateritic soil + 30% marine clay	12.97	0.96	24.66	1.60	0.99
Lateritic soil + 10% bentonite	48.64	0.98	55.59	1.28	0.99
Lateritic soil +20% bentonite	5.10	0.99	12.27	1.55	0.98
Lateritic soil +30% bentonite	8.15	0.99	14.92	1.42	0.97

k_d - Partition coefficient; k_F - Freundlich capacity factor; n - an empirical constant in aqueous solution. Although the value of n shows a favorable adsorption, the R^2 values in Table 3 indicate that test results are not fit well for Freundlich model for high concentration Cr (VI) solutions.

Table 3 Frendlich adsorption parameters all test data (0.5-100mg/L)

Soil	Freundlich isotherm		
	k_F (mg/kg)	n (kg/L)	R^2
Lateritic soil	6.14	1.51	0.892
Lateritic soil + 10% BC soil	4.94	1.51	0.982
Lateritic soil + 20% BC soil	3.97	1.66	0.981
Lateritic soil + 30% BC soil	4.95	1.76	0.836
Lateritic soil + 10% marine clay	9.97	2.83	0.754
Lateritic soil + 20% marine clay	22.23	2.09	0.938
Lateritic soil + 30% marine clay	40.36	1.81	0.941
Lateritic soil + 10% bentonite	12.55	1.58	0.988
Lateritic soil + 20% bentonite	6.14	1.51	0.892
Lateritic soil + 30% bentonite	4.94	1.51	0.982

k_F - Freundlich capacity factor; n - an empirical constant