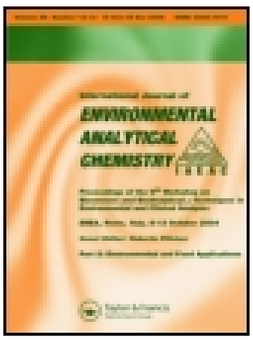

International Journal of Environmental Analytical Chemistry

ISSN: (Print) (Online) Journal homepage: https://www.tandfonline.com/loi/geac20

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To cite this article: V. Sathya, M. Sinduja, P. Kalpana, M. Maheswari, M.R. Ramasubramaniyan & S. Mahimairaja (2021): Strategic study of adsorption and desorption of chromium on vertisols and its implication in developing an effective remediation technology, International Journal of Environmental Analytical Chemistry, DOI: 10.1080/03067319.2021.1940985

To link to this article: https://doi.org/10.1080/03067319.2021.1940985

Published online: 23 Jun 2021.

INTERNATIONAL JOURNAL OF ENVIRONMENTAL ANALYTICAL CHEMISTRY
https://doi.org/10.1080/03067319.2021.1940985

Strategic study of adsorption and desorption of chromium on vertisols and its implication in developing an effective remediation technology

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ABSTRACT

Chrome tanning in India leads to severe contamination of soil and water. Due to the indiscriminate disposal of effluent from tanneries, several thousand hectares of agricultural lands have been degraded over several decades in Tamil Nadu. Several researchers were involved in developing remediation technologies for chromium contamination.Adsorption and desorption of Cr on soil determine the mobility of Cr and its environmental impact. There are several studies regarding the adsorption/desorption of Cr. A strategic study regarding the mechanism of adsorption/desorption of Cr to the context of remediation technologies is essential. A better understanding of the adsorption and desorption mechanism of Cr aids in developing a suitable remediation strategy. Therefore, in the present investigation, the adsorption Cr on soil and some critical factors affecting the adsorption were studied. The Cr adsorption on soil recorded maximum and was best illustrated by the Freundlich isotherm (R^2 = 0.994) than the Langmuir isotherm (R^2 = 0.841). Amongst the bio-amendments, biochar was found to enhance the adsorption of Cr on the soil. The co-contaminants such as cadmium and lead significantly reduced the adsorption of Cr. Further, a desorption study was carried out with different desorption agents such as NaOH, $CH_3COONH_4$and EDTA. It was found that NaOH exhibited higher desorption efficiency than the other agents. The results reveal that adsorption and desorption mechanisms aids in predicting the toxicity of Cr. This study also shows that the soil pH, bio-amendments, and co-contaminants in the contaminated soil may play a significant role in the remediation process.

ARTICLE HISTORY
Received 5 March 2021
Accepted 3 June 2021

KEYWORDS
Chromium; remediation; bioavailability; adsorption; desorption

1. Introduction

The tannery industry is one of the profitable sectors in India, which makes enormous amounts of money of foreign exchange earnings to the country. More than 70% of India's

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tanning industries are located in Tamil Nadu, especially in Vellore District. Most of the tanneries in India adopted chrome tanning instead of the vegetable tanning process because of its rapidity, low costs, quality, and stability of the produced leather [1]. Apart from the profit and employment benefits, leather industries discharge 20–40% unused toxic heavy metal, namely chromium (Cr) containing wastewater in into the sewage system, which has degraded soil and water quality and severe threat to the environment and human health [2,3]. According to [4], the worldwide discharge of 52,600 tonnes of Cr from 1762 industries has created considerable risk to the environment and health due to direct contamination and leaching of Cr from the soil during raining and subsequently waterlogged circumstances.

The solubility and bioavailability of the heavy metals are determined through various physico-chemical and biological mechanisms occurring in contaminated soil. However, several factors are restraining the solubility of metals in soil. Adsorption/desorption is the prime factor determining the mobility and further environmental implications of metals [5]. The quantity of surface negative charges of the soil primarily determines the ability of adsorption by the soil. Hence, a study focusing on the patterns of adsorption of heavy metals on soil particles is vital to assess the mobility of heavy metals from liquid to a solid phase [6]. The soil particles have a considerable role in the retention and migration of Cr ions to the groundwater [7]. The adsorption/desorption of anionic Cr in the soil occurred by a specific/non-specific adsorption process and was influenced by soil properties such as pH, inorganic minerals, electrolyte species, etc [8].

The adsorption pattern of Cr slows the state equilibrium of Cr, leads to the poor fitting performance of the kinetic data [9]. The reaction of reducing hexavalent Cr to trivalent Cr is one of the prerequisites to remediate the contaminated soil and water due to its less toxicity and mobility [10]. The chemical properties of soil and the composition of the aqueous soil phase decide the dynamic equilibrium between Cr in the solid and aqueous soil phase [11]. Desorption is a phenomenon or process in which any of the material that has been adsorbed is released back to the soil. Physical bonding, ion exchange, or a mixture of both may bind a solute to some adsorbent. In some instances, the loosely bound solute can be readily desorbed when using distilled water, and the adsorption process is physical bonding. If the adsorption is chemical bonding, ion exchange, or both, stronger desorbents such as acid or alkali solution may influence the adsorption [12].

The disposal of Cr encumbered adsorbents in the ecosystem is unsafe. The desorbed metals from adsorbents paved the way for recovering the metals and reducing the problem related to metal disposal contained adsorbents [13]. Further, for the adsorption to be more feasible and applied practically, it is vital to observe the recuperation of the adsorbents through the restoration process by treating the utilised adsorbent substance with appropriate extractants such as Nitric acid, Sodium hydroxide, Ammonium acetate, Ethylenediaminetetraacetic acid (EDTA) [14]. Therefore, in our study, the Cr adsorption/desorption pattern was examined through a series of laboratory batch experiments. Further, the impact of different soil properties such as pH of the soil, organic matter content of the soil, and co-contaminants in soil on Cr adsorption was studied to develop effective remediation technology for Cr contaminated soil.

2. Materials and methods

A sequence of laboratory experiments of different batches was carried to investigate the adsorption patterns of Cr on the soil. The soil used for the experiment was collected from a farm of Centre for Rural Development, National Agro Foundation, Illedu, Chengalpattu District, Tamil Nadu, India. The initial characteristics of the soil were examined and depicted in Table 1. The soil type is clay loam, and the USDA classification of soil is vertisol. It has a neutral pH with an electrical conductivity (EC) of 0.542 dSm^{-1}. The organic matter of soil was 0.96%. The macro and secondary nutrients contents of the soil ranged from medium to high. The micronutrient status was found to be lower. The soil had a high (22.4 Cmol (p+) kg^{-1}) cation exchange capacity (CEC). The concentration of Cr in the experimental soil was found below the detectable limit (Bdl).

2.1. Experiment methods to assess the adsorption of Cr in soil

Firstly, the Cr adsorption was assessed at various ranges of time from 30 minutes to 48 hours using a constant Cr (100 mg L^{-1}) constant concentration. Finally, it was recorded that the maximum of adsorption happened within 12 hours. Then, a batch experiment was conducted to obtain the adsorption isotherm with various concentrations of Cr ranged from 0 to 100 mg L^{-1}, and the period was constant (12 hours). One gram of soil was mixed with 25 ml of Cr solutions in polypropylene tubes and subjected to shaking on an end-over-end shaker for the equilibrium time of 12 hours at room temperature. Finally, the batch of samples was centrifuged at the speed of 8000 rpm for 10 minutes and then filtered. Then the final extract solutions were analysed for the equilibrium concentration of Cr using the Atomic Absorption Spectrophotometer (VARIAN AA220). The difference between the initial and final levels of Cr in solutions was determined, and then the amount of Cr adsorbed per unit weight of soil was computed. The adsorption data

Table 1. Characteristics of experimental soil.

Parameters	Unit	Values
Soil Type	-	Clay loam
USDA classification	-	Vertisols
Bulk density	(Mg m^{-3})	1.31
pH	-	7.30
Electrical Conductivity	dSm^{-1}	0.542
Organic matter	%	0.96
Nitrate Nitrogen	mg kg^{-1}	63.7
Available Phosphorus	mg kg^{-1}	23.72
Exchangeable Potassium	mg kg^{-1}	428
Exchangeable Calcium	mg kg^{-1}	2601
Exchangeable Magnesium	mg kg^{-1}	815
Available sulfur	mg kg^{-1}	44.0
Exchangeable Sodium	mg kg^{-1}	346
Available Zinc	mg kg^{-1}	0.96
Available Manganese	mg kg^{-1}	9.38
Available Iron	mg kg^{-1}	4.55
Available Copper	mg kg^{-1}	2.34
Available Boron	mg kg^{-1}	0.5
Total Chromium	mg kg^{-1}	Bdl
Cation exchange capacity	Cmol (p+) kg^{-1}	22.40

were fitted in Equation 1 and 2 to find out the best of typical isotherms such as Freundlich and Langmuir isotherms, respectively

$$\left(\frac{x}{m}\right) = K_f Ce^n \quad \text{(Eq.1)}$$

where (x/m) is amount adsorbed (mg kg^{-1}), Kf is Freundlich distribution coefficient, Ce is equilibrium solution concentration (mg/L), and 'n' is equilibrium constant (dimensionless).

$$Y = \frac{MbC}{(1 + bC)} \quad \text{(Eq.2)}$$

Where Y = amount adsorbed (mg kg^{-1}), C = equilibrium solution concentration (mg L^{-1}), and b is the Langmuir constant, related adsorption energy.

2.2. Factors affecting adsorption of Cr on soil

The adsorption of Cr on soil is affected by various factors like soil pH, organic matter content, and level of contaminants in soil. The influence of pH on the adsorption of Cr was assessed between pH 5 and 9. The solutionpH was attuned to a necessary pH range such as 5, 6, 7, 8, and 9 using 0.1 M HCl or 0.1 M NaOH. The impact of soil organic matter on Cr adsorption was inspected with different bio-amendments such as farmyard manure (FYM), poultry manure (PM), composted press mud (CPM), and biochar (BC) at a rate equivalent to 0.5 g g^{-1} of soil based on a preliminary experiment conducted. The bio-amendments were processed and sieved through a 2 mm sieve and evaluated for a few vital characteristics (Table 2). The effect of co-contaminants typically occurs in the soil on the adsorption of Cr was observed. The dominant metals that occur together with Cr in the soil are Cd and Pb. Therefore, these two metals were selected for investigation. The difference between the initial concentration and final equilibrium solution concentrations was determined for each experiment to compute the quantity of Cr adsorbed per unit weight of soil.

2.3. Experiment methods to assess the desorption of Cr in soil

Desorption characteristics of Cr present in the soil were studied by conducting batch experiments using different desorption agents such as NaOH, CH_3COONH_4, and EDTA. Similar to the adsorption studies, Cr desorption was also assessed over a period of 30 mins to 48 hours with a constant concentration of Cr (100 mg L^{-1}). One gram of soil was mixed

Table 2. Physio-chemical characteristics of bio-amendments used.

Parameters	Unit	Poultry manure	FYM	Biochar	Pressmud compost
pH		6.23	7.25	7.86	6.27
EC	dSm^{-1}	2.35	3.08	3.19	2.28
Organic Carbon	%	20.42	24.12	5.16	16.80
Total Nitrogen	%	2.75	1.85	1.19	2.29
Total Phosphorous	%	1.44	7.54	10.35	10.18
Total Potassium	%	0.86	0.13	0.76	0.62
Nickel	mg kg^{-1}	16.26	16.01	18.07	13.50
Lead	mg kg^{-1}	24.07	19.65	24.00	21.05
Chromium	mg kg^{-1}	0.47	BDL	BDL	0.33
Cadmium	mg kg^{-1}	3.14	3.69	3.86	3.07

with 25 ml of Cr solutions in polypropylene tubes and subjected to shaking on an end-over-end shaker for the equilibrium time of 30 mins at room temperature. The samples were centrifuged at 8000 rpm for 10 minutes and filtered using Whatman No. 40 filter paper. The supernatant was discarded, and the filtrate was collected. Then, 25 ml of desorption agents [0.1 M of NaOH, CH_3COONH_4, and EDTA] was added to the Cr adsorbed soil (filtrate). The desorption at a range of equilibrium period (0, 0.5, 1, 2, 5, 8, 12, 24, 36, 48 hours) was examined. Further, the desorption of Cr has been calculated by adding different concentrations of Cr between 0 and 100 mg kg-1 to the soil, and the desorption agents were added to the Cr adsorbed soil to find out the maximum desorption rate of Cr from the soil. The difference between the initial and final levels of Cr in solutions was determined.

3. Results and discussion

It is widely acknowledged that adsorption/desorption is the primary mechanism that controls the bioavailability and mobility of Cr in contaminated soil. Based on the patterns of adsorption of Cr, a suitable remediation technology for Cr contaminated soil could be developed. Therefore, a sequence of laboratory experiments was conducted to examine the Cr adsorption/desorption on the experimental soil and the factors affecting the adsorption of Cr on the soil. The results are discussed below.

3.1. Adsorption of chromium on soil as a function of equilibrium period

The equilibrium period when the maximum amount of Cr is adsorbed and retained by soil particles was determined. The result confirmed that the Cr adsorption progressively amplified with time and attained a maximum (74.66 mg kg^{-1}) at 12 hours with a constant input concentration of Cr (100 mg kg^{-1}) and continued up to 48 hrs (Figure 1). It was showed that Cr adsorption attained steady-state equilibrium within 12 hours. Therefore, in subsequent experiments, an equilibrium time of 12 hours was followed. The rapid

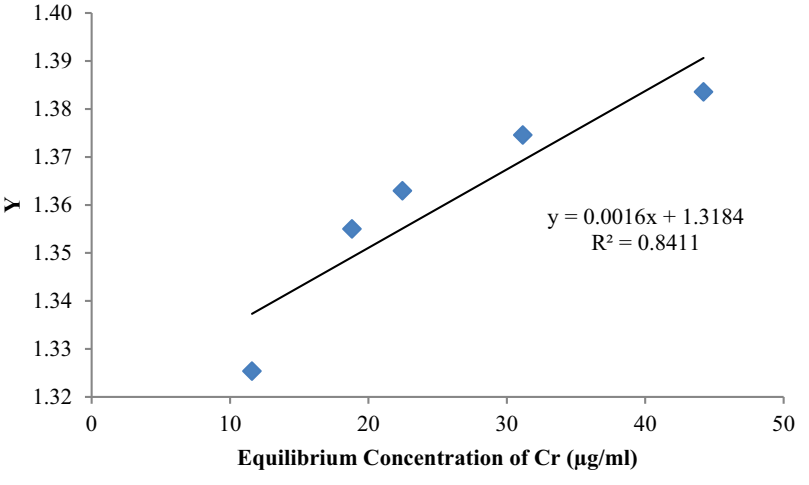

Figure 1. Langmuir isotherm.

adsorption during the initial hours is attributed to higher soil surface area availability since experimental soil has a higher CEC range. Once the Cr ions occupy the active sites, the adsorption remains constant without any changes. Thus, it has been attained an equilibrium state at 12 hrs [15]. The investigation of changes in adsorption patterns explains the rate of solute uptake, and obviously, it influences the residence time adsorbate uptake at the soil-aqueous interface [16]. The adsorption and Cr withholding capacity of soil establish its persistence, responses, changes, and mobility [12,17].

3.2. Adsorption of chromium on soil as a function of equilibrium concentration

The most significant factor that controls the mechanism of adsorption is the initial amount of metal present in the soil. Therefore, the effect of various concentrations (0, 20, 40, 60, 80, and 100 mg L^{-1}) of Cr on adsorption was examined. The result showed that the Cr adsorption was significantly controlled by the initial quantity of Cr in the soil. The equilibrium concentration of Cr varied from 11.59 to 44.22 mg L^{-1} (Figure 3). The range of Cr adsorbed on soil varied from 8.41 to 55.78 mg kg^{-1}. While the initial concentration increased, the adsorption was increased progressively, and the maximum adsorption (55.78 mg kg^{-1}) was observed at the concentration of 100 mg L^{-1}. The results indicate that the Cr adsorption was increased when the amount of Cr present in the soil increased. It might be due to the availability of exchange sites in the soil, which paves the way for enhanced adsorption of Cr on the soil's surface [17–19]. The increased amount of initial Cr in the soil can act as a driving force to manage all mass transfer resistances for a liquid to the solid phase of soil [20].

3.3. Adsorption isotherm

The results recorded in batch experiments were examined by fitting into the typical isotherms such as Langmuir and Freundlich isotherms. The values of the correlation coefficient (R2) were compared to get the best fit of isotherms. The adsorption data of the two isotherms (Equations (1) and (2)) concluded from graphical representations (Figures 1 and 2). The values of R^2 suggest that the Freundlich isotherm gave the highest accordance with experimental data.

The Freundlich isotherm was also applied to the biosorption data. This empirical model explains the heterogeneous adsorption of molecules in the soil or adsorbent. It is also proposed for the exponential decrease of energy involved in the adsorption after completing the adsorption process with the soil surface or adsorbent. The 'n' value denotes the rate and nature of non-linearity between adsorption and solution concentration in the Freundlich isotherm. If 'n' = 1, it represents the linear adsorption; if 'n' > 1, then it is a physical adsorption process; and if 'n' <1, it is a chemical adsorption process. The obtained value for 'n' for Cr ions removal was found to be 1.406, indicating physical adsorption processes [21].

3.4. Influence of soil pH on adsorption of chromium

The pH of the solution is the significant influencing factor in the adsorption study because it dramatically affects the hydrogen or hydroxyl ions of soil and the surface charge of the

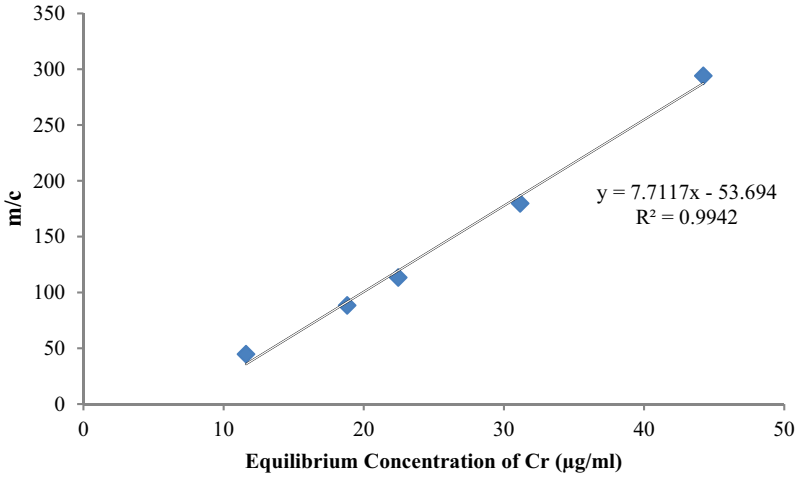

Figure 2. Freundlich isotherm.

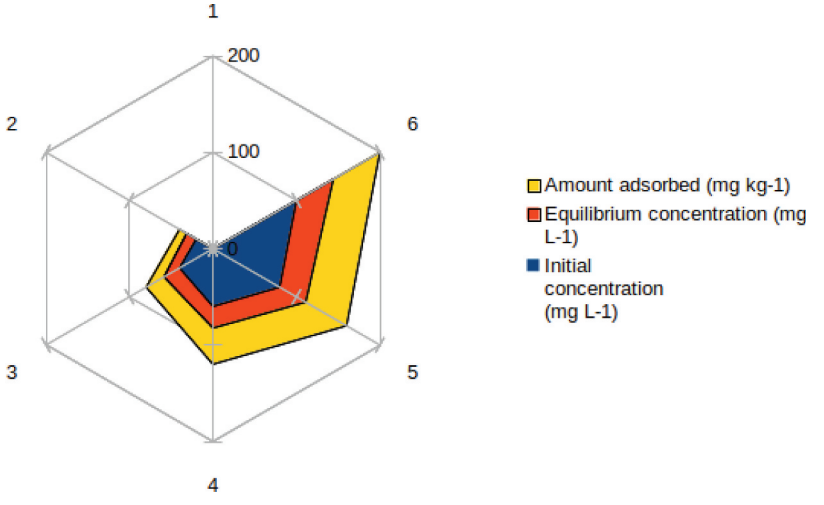

Figure 3. Adsorption of Chromium on the soil as a function of the equilibrium concentration.

adsorbent under a working solution [22]. Hence, the study on pH impact on adsorption is essential for establishing Cr under normal soil conditions. In this study, initial soil pH was adjusted to have a range of values from 5.0 to 9.0, and its impact of Cr adsorption was studied. The quantity of Cr adsorbed at various pH levels ranged between 73.93 and 99.65 mg kg^{-1}. When pH increased, there was a significant decrease in the adsorption of Cr. The uppermost quantity of Cr (99.65 mg kg^{-1}) adsorbed on soil was observed at pH 5 (Figure 4). The influence of pH on the adsorption of heavy metal may widely be linked with the functional group types present in the soil or adsorbent and its ionic state and the chemistry of metals present in the solution [23]. The Cr ions present in various forms like $Cr_2O_7^{2-}$, $HCrO^{4-}$, $Cr_3O_{10}^{2-}$, $Cr_4O_{13}^{2-}$ but $HCrO_4$dominates all other forms at lower pH ranges. If solution pH increases, the other species, such as CrO_4^{2-} and $Cr_2O_7^{2-}$ will

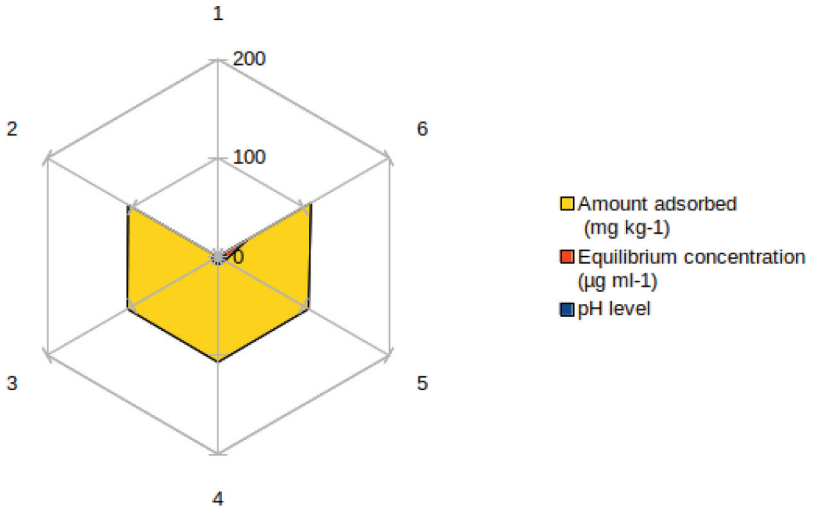

Figure 4. Effect of soil pH on adsorption of Chromium.

dominate the process. At acidic pH level (1 to 6), the amount of H^+ ions on the soil surface and the adsorbent surface will be increased, resulting in the strong electrostatic attraction between the positively charged soil or adsorbent surface and various chromate ions. Hence, there will be a rise in the adsorption of Cr at lower pH levels. There is an assumption that hexavalent Cr might be reduced to trivalent Cr under acidic pH conditions. Conversely, hexavalent Cr ions present as an oxo-anionic form in a highly acidic medium [24,25].

Similarly, at higher pH levels >6, there might be a competition between anions such as $CrO_4^{2-}$and OH^- to obtain adsorption sites on the surface of soil or adsorbent where OH^- will dominate the process. Hence, the adsorption rate of Cr at a higher pH level decreases [26–28].

3.5. Influence of soil organic matter on adsorption of chromium

The soil organic matter plays a significant role in Cr adsorption on the soil. Various bio-amendments such as farmyard manure, poultry manure, composted press mud, and biochar were used at a rate of 0.5 g g^{-1} of soil and a constant Cr concentration of 100 mg $kg^{-1}$to evaluate the impact of soil organic matter on Cr adsorption. The rate of adsorption of Cr due to bio-amendments ranged between 5.94 and 23.06 mg kg^{-1} (Figure 5). In general, the application of amendments significantly influenced the Cr adsorption. The highest amount of Cr adsorbed was observed in soil amended with biochar, while the lowest in the control soil. It could be ascribed to the functional groups there in the biochar, larger specific surface area, and higher porosity of the soil, which paved the way for a higher rate of Cr adsorption [29,30]. The Cr oxyanions present in the solution will react with the electron donors such as bio-amendments such as COOH, OH^-, and NH_2. After the redox reaction occurs, the hexavalent Cr ions are reduced to trivalent Cr cations, which are generally less toxic. The textural property of an amendment and its pore filling adsorption mechanism have a significant role in influence the adsorption

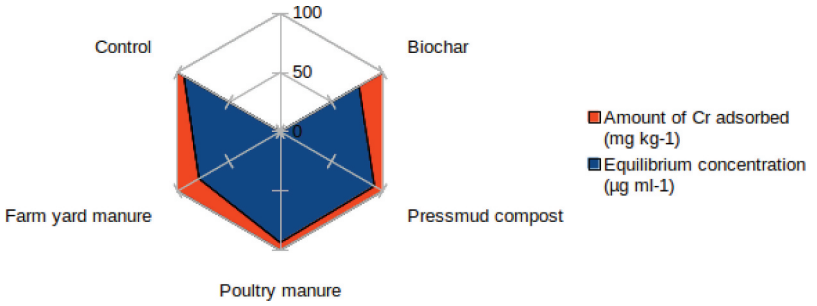

Figure 5. Effect of bio-amendments on adsorption of Chromium.

capacity to different contaminants such as total Cr, Cr (VI), and Cr (III) [31,32]. The adsorption capacity varies with the type of bio-amendments used, and it is ascribed to the amount of functional group present in each bio-amendments [33].

3.6. Influence of co-contaminants on adsorption of chromium

The influence of Cd and Pb at a constant concentration (100 mg kg^{-1}) on different concentration ranges (0 to 100 mg kg^{-1}) of Cr was examined. The occurrence of various co-contaminants affected the Cr adsorption on soil (Figures 6 and 7) considerably. When Cd occurred as a co-contaminant, the equilibrium concentration ranged between 1.77 and 15.87 mg kg^{-1}, whereas Pb was a co contaminant, the range of equilibrium concentration of Cr was recorded between 4.06 and 15.99 mg kg^{-1}. Chromium adsorption was found to be decreased significantly in the presence of Cd and Pb. This may be due to the adsorption capacity of Cr under competitive conditions. The rate of Cr adsorption was elevated at the beginning due to the surface area and availability of sorption sites for Cr while comparing with Cd and Pb. Relatively same findings were observed by [34]. Several

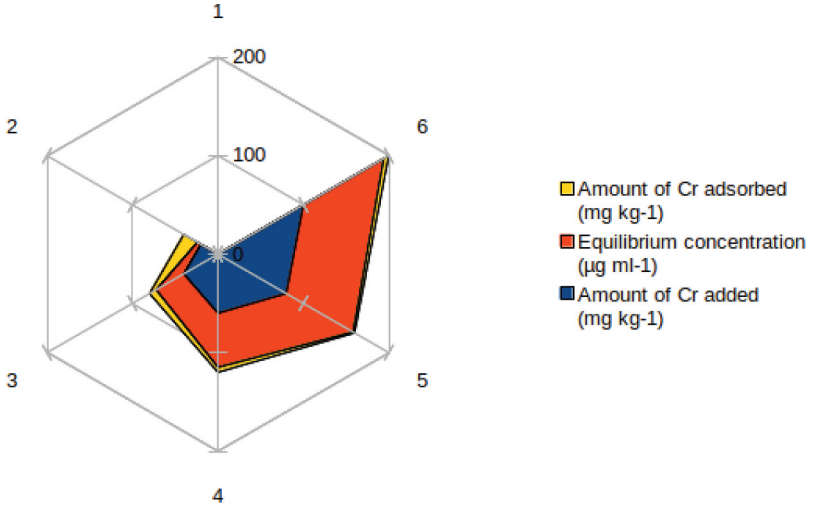

Figure 6. Effect of co-contaminant (Cd) on adsorption of Chromium.

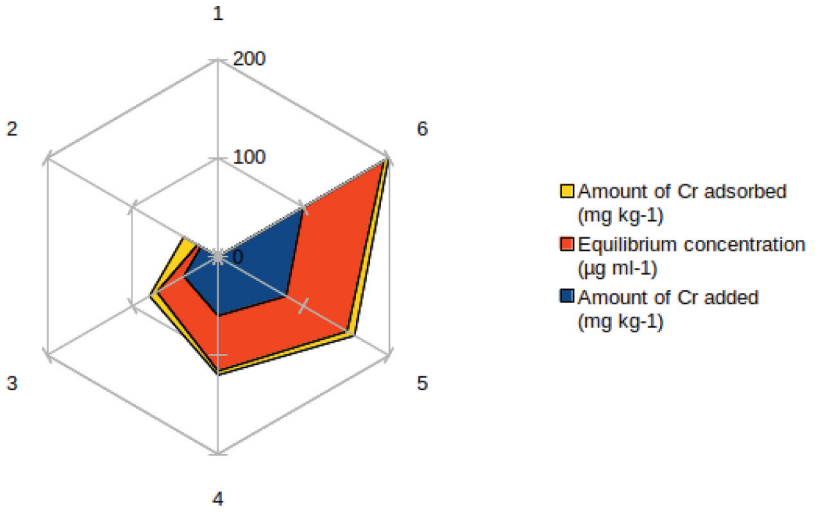

Figure 7. Effect of co-contaminant (Pb) on adsorption of Chromium.

factors such as pH of the soil, residence time, initial metal concentration, and temperature decide the capacity of adsorption for each metal [35]. It is also observed that the adsorption capacity of heavy metals in a binary system or multi-metal system will be much less than in the metals single metal system [36].

3.7. Desorption of chromium on soil as a function of equilibrium period

When compared with the adsorption process, the study of the desorption process is relatively significant. However, desorption is a significant factor that affects the migration of pollutants in the soil, and desorption is an inverse process of adsorption [37]. The desorption study of Cr was carried out with 0.1 M of NaOH, CH_3COONH_4, and EDTA solutions as a function of the equilibrium period. The result revealed that the Cr desorption progressively intensified with time and attained a maximum (2.17 mg kg^{-1}) at 12 hours with a constant input concentration of Cr (100 mg kg^{-1}) (Figure 8). Similar to the adsorption studies, rapid desorption was observed in the initial hours. This may be due to the higher CEC range of the soil [38]. It was observed that the amount of Cr adsorbed in the soil ranged between 21.82 and 74.66 mg kg^{-1}. Further, when these desorbing agents were added, and the desorption of Cr have reached upto 100%. This indicates that more Cr has been desorbed from the soil to the solution while using NaOH as an extractant. On comparing the effectiveness of extractants, NaOH exhibited the highest desorption percentage of about 100%, whereas CH_3COONH_4 exhibited the highest desorption percentage of 41.50%, and EDTA exhibited the highest desorption percentage of 95.24%.

3.8. Desorption of chromium on soil as a function of equilibrium concentration

Desorption of Chromium on soil was conducted with different extractants such as NaOH, CH_3COONH_4, and EDTA. Different concentrations of Cr were added to the soil, and the desorption agents were added to the Cr adsorbed to the soil. It is apparent from Figure 9

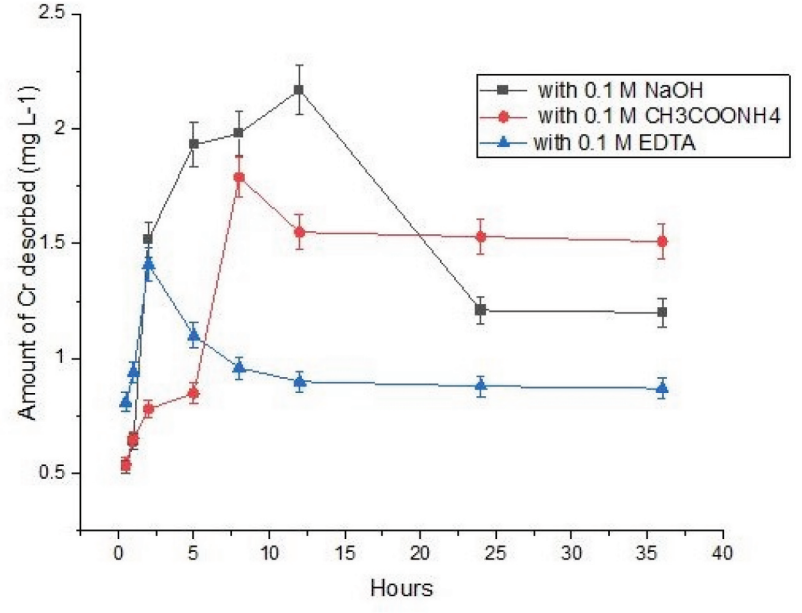

Figure 8. Desorption of Chromium on soils as a function of equilibrium period.

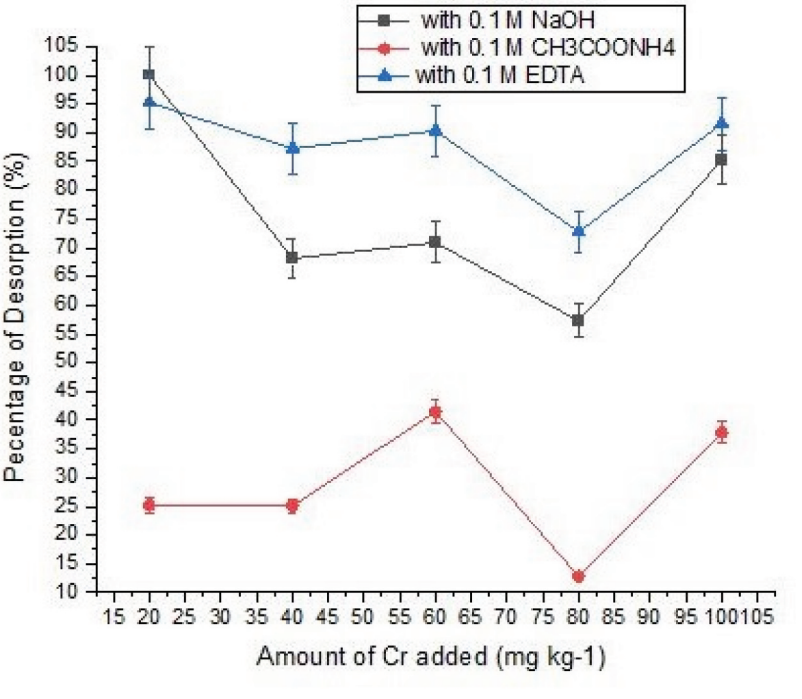

Figure 9. Desorption of Chromium on soils as a function of the equilibrium concentration.

that as the initial concentration of Chromium increases, percent desorption also increases. On increasing the adsorbed concentration from 8.41 to 55.78 mg kg-1, desorption of Chromium varies from 12.94% to 100% with different extractants. Desorption studies aids in elucidating the nature of adsorption. Usually, the adsorption of solute on an adsorbent can be physical bonding, ion exchange, or a combination of both methods. If the adsorption occurs by physical bonding, then the loosely bound solute can be easily desorbed by using distilled water in most cases. But, if the adsorption process is due to chemical bonding or ion exchange or a combination of both, desorption is affected by strong desorbents like acids or alkali solutions [39]. Considering all these effects, the results of desorption of Cr by desorbents such as NaOH, CH_3COONH_4, and EDTA suggest either chemisorption or ion exchange as the possible mechanism of Chromium adsorption onto the soil [40].

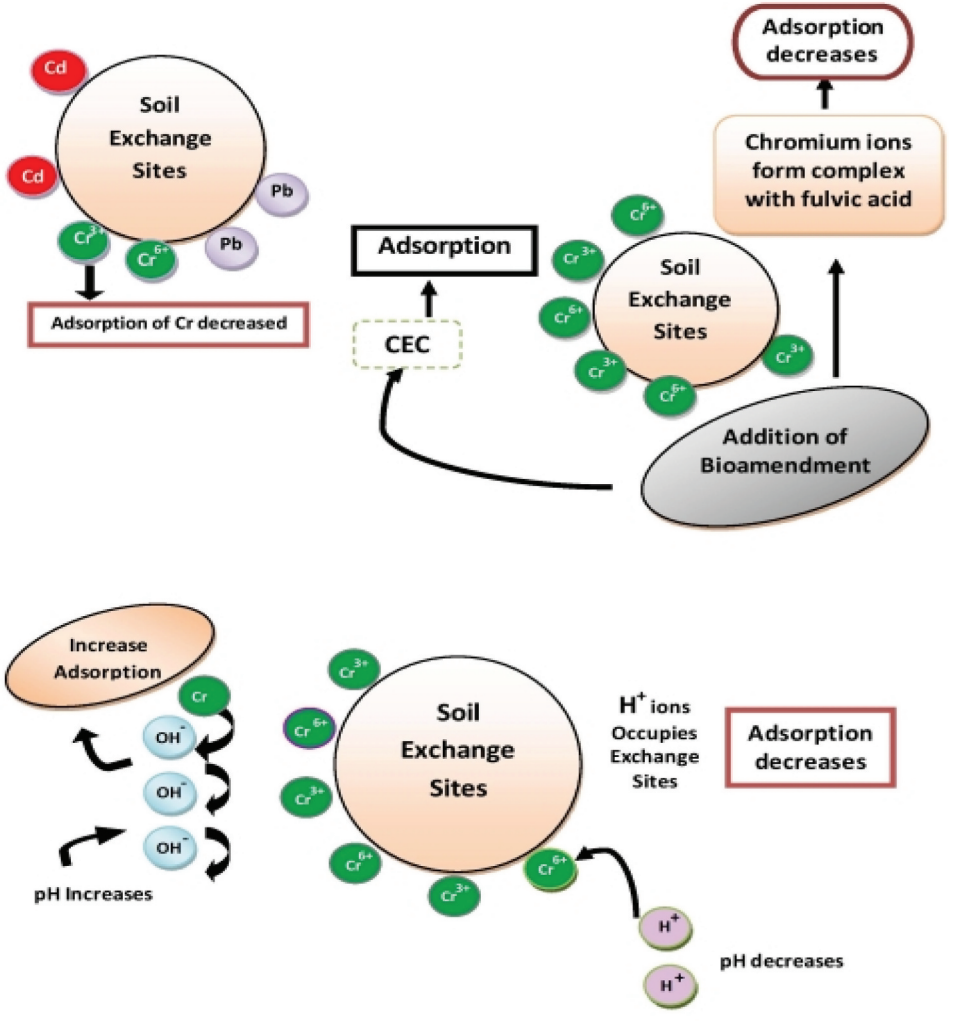

Figure 10. Mechanism of Adsorption of Cr on soil with respect to different pH, Bio-amendments and Co-Contaminants.

The following inference was made from the results of desorption studies: The highest percent desorption of Chromium with NaOH indicates that the phenomenon is consistent with the results observed for the effect of pH of solution for adsorption, which goes well with the study of [41]. At the high strength of NaOH solution, the abundance of hydroxide ions (-OH) increased the competitiveness between Chromium anions and hydroxide ions for the adsorption at the soil's surface, so detachment of the Chromium anions was increased from the surface of the soil. Among the NaOH, CH_3COONH_4, and EDTA desorbents, NaOH is the best extractant because it can form water-soluble complexes. The obtained results are in agreement with the previous studies [39,41]. The overall Mechanism of Adsorption of Cr on soil with respect to different pH, Bio-amendments and Co-Contaminants (Figure 10).

4. Conclusion

Adsorption is a crucial mechanism that governs the transportation and conversion of Cr in soil and its environmental impact. This study reveals that the percentage removal of Cr ions depended on the different factors, such as the initial concentration of Cr and the equilibrium period. The adsorption process followed the Langmuir isotherm, and it reveals that the adsorption of Cr is mainly a physical process. There is a pronounced influence in the adsorption of Cr by soil properties such as pH of the soil, organic matter content of the soil, and co-contaminants that occur in the soil. The remediation strategies like immobilisation or mobilisation of metal ions and subsequent removal by phytoremediation may be selected based on the adsorption patterns of metal and other soil properties that influence the mobility of the metals. Based on the present adsorption batch experiments conducted, it may be concluded that if the contaminated soil is acidic in nature, the immobilisation technique using bio-amendments such as biochar and FYM could be used way more significantly adsorption of Cr in soil. The dosage of bio-amendments may also be appropriately calculated based on the other heavy metals present in the contaminated soil since they have significant implications on the Cr adsorption and mobility.

Further, desorption studies reveal that the desorption increases on increasing initial Chromium concentration due to the physical bonding of Chromium onto the soil. Further, NaOH solution extracted more Cr and found effective than other extractants. With this study, it is possible to predict the mobility and transformation of Cr. Thus, it helps in planning control measures to eliminate the toxic effects of Cr present in the soil.

Acknowledgments

The author would like to thank DST-SERB, Government of India, New Delhi under Early Career Research AwardGrant N. ECR/2016/000971

Data Availability Statement

The authors confirm that the data supporting the findings of this study are available within the article.

Disclosure of potential conflicts of interest

We, the authors of this manuscript, certify that we have NO affiliations with or involvement in any organization or entity with any financial or non-financial interests in the subject matter or materials discussed in this manuscript.

Funding

This work was supported by DST-SERB, Government of India, New Delhi under Early Career Research AwardGrant No. (ECR/2016/000971)

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