

# Batch Chromium(VI), Cadmium(II) and Lead(II) Removal from Aqueous Solutions by Horticultural Peat

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**Abstract** The selectivity and uptake capacity of horticultural peat available in Romania was evaluated with respect to the removal of Cd(II), Cr(VI) and Pb(II) ions from aqueous solution. The kinetics, sorption capacities, selectivity and pH dependence of sorption were determined. The influence of metal concentration in solution is discussed in the terms of Langmuir and Freundlich isotherm and constants. Sorption capacities increased with increasing metal concentration in solution. For solutions containing 300 mg/l of metal, the observed uptake capacities were 20 mg Cd(II)/g peat, 15 mg Cr(VI)/g peat and 30 mg Pb(II)/g peat. The study proved that horticultural peat is a suitable material for the removal of the studied heavy metal ions from aqueous solutions, achieving removal efficiencies higher than 90%, and could be considered as a potential material for treating effluent polluted with Cd(II), Cr(VI) and Pb(II) ions.

**Keywords** Adsorption isotherms · Adsorption kinetics · Cadmium · Chromium · Lead · Peat · pH effect

## 1 Introduction

Many industries discharge wastewaters containing heavy metals such as chromium, cadmium, lead, among others. Since the early 1970s, however, there has been growing concern over the effect of heavy metals on humans and aquatic ecosystems.

Legislation on the limit concentrations for toxic metal ions discharged in wastewater is now very stringent in Romania. The technology normally employed for metal removal in industrial applications has been precipitation. The motivation for a change away from precipitation technologies in metal control applications derives from the fact that precipitation can only reduce the concentration of dissolved metals to the solubility product level, which often does not comply with stringent discharge permission standards and calls for further polishing stages.

The management of metal-contaminated sludge is costly and it is very difficult to treat wastewater containing low concentrations of heavy metals.

Since adsorption is one of the more effective methods for removing heavy metals from wastewater, an inexpensive and more easily available adsorbent would make the removal of heavy metals from wastewaters by adsorption an economically viable alternative.

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Over the last four decades, peat has gained recognition as an agent for water pollution control. Peat is defined as a fibrous material of organic matter in various stages of decomposition, dark in color and of a spongy consistency, being a non-consolidated hydrophilic, carbonaceous sediment (Viraraghavan and Rao 1993). It may also be defined as unconsolidated, hydrophilic carbonaceous sediment formed by the accumulation of partially fragmented, decomposed and commonly heterogeneous plant remains which retain more than 75% inherent moisture and less than 12% mineral matter in saturated natural deposits. Peat constituents, especially lignin, contain polar functionalities, such as alcohols, aldehydes, ketones and acids, which can be involved in chemical bonding of cations (Couillard 1994).

The abundance and the availability of peat make it economically feasible. In its raw form, however, physical characteristics such as low mechanical strength, high affinity for water, poor chemical stability and a tendency to shrink and swell (Coupal and Lalancette 1976) must also be taken into consideration as restrictive features in its industrial application.

Due to the very polar nature of this material, the specific adsorption for dissolved solids, such as metals, is reported to be quite high. Several studies have established the potential of peat to capture dissolved metals, nutrients, suspended solids, organic matter, oil, etc. (Malterer et al. 1996).

Peat has a greater cation exchange capacity than mineral soil and inorganic soil. Lalancette and Coupal (1972) found peat to be efficient at removing Hg from waters. Bunzl et al. (1976) reported the efficiency of peat in the adsorption of Pb(II), Cu(II), Cd(II) and Zn(II). Chaney and Hundemann (1979) reported sphagnum peat to be effective in removing cadmium from simulated electroplating wastewater. Sharma and Forster (1993) found sphagnum peat to be effective in removing chromium(VI) from aqueous solution. Gardea-Torresdey et al. (1996) found peat to be efficient for Cu(II) uptake and Ho et al. (1995, 2002), Ho and McKay (2004) studied batch nickel, copper and lead sorption onto peat.

The aim of this study was to evaluate the effectiveness of locally available horticultural peat for the removal of cadmium(II), chromium(VI) and lead(II) ions from aqueous solutions. These cations are very toxic and, even in small quantities, could be detrimental to most forms of life. Cadmium can damage the kidneys, lead affects red blood cells and the nervous

system, and exposure to hexavalent chromium can produce acute poisoning (Brown et al. 2000).

Laboratory batch experiments were conducted to evaluate the adsorption capacity of peat towards Cd(II), Cr(VI) and Pb(II), studying the effect of contact time and pH.

The influence of metal concentration is discussed in the terms of Langmuir and Freundlich isotherms.

## 2 Materials and Methods

### 2.1 Materials

A standard pack (50 l) of horticultural peat commonly available in Romania, produced by HOGO TUINPRODUKTEN BV Holland, was used for the study. The raw peat was air dried at room temperature (25°C) and then screened through a 14-mesh sieve to remove any large non-peat solids. The resulting material is uniform and appropriate for sorption tests.

Aqueous solutions containing heavy metals were prepared using analytical reagents and distilled water. Stock solutions for each metal (containing 1,000 mg metal/l) were prepared using “analytical grade” potassium dichromate, cadmium sulphate and lead nitrate.

All working solutions were prepared by diluting the stock solution with distilled water. pH adjustment was performed using sulphuric or nitric acid and sodium hydroxide 0.1 N solutions.

The characteristics of the horticultural peat employed in this research study are presented in Table 1.

### 2.2 Batch Kinetic Studies

For each of the adsorption kinetic test, 1 g peat was thoroughly mixed with 100 ml solution containing 100 mg/l of Cd(II), Cr(VI) or Pb(II). Peat suspensions were shaken on a rotary shaker at 100 rpm in 250 ml conical flasks at room temperature to determine the equilibrium time, defined as the contact time required for the concentration of heavy metal in solution to reach an equilibrium value, for each peat–metal system. At the end of each one hour period, the suspension from each jar was filtered through a 0.45 µm Millipore membrane filter and metals were determined in the solution by atomic absorption spectroscopy, using a Karl Zeiss AAS 30 instrument. All batch kinetic studies were replicated.

**Table 1** Characteristics of horticultural peat employed

Characteristic	Value	Anions	Extract (mmol/l)	Cations	Extract (mmol/l)
Organic materials	90%	Cl <sup>-</sup>	0.3	NH <sub>4</sub> <sup>+</sup>	1.4
Ash	20%	SO <sub>4</sub> <sup>2-</sup>	0.9	K <sup>+</sup>	0.8
Humidity max.	50%	PO <sub>4</sub> <sup>3-</sup>	0.7	Ca <sup>2+</sup>	0.8
Conductivity	1,400 μS/cm	NO <sub>3</sub> <sup>-</sup>	1.4	Mg <sup>2+</sup>	0.4
pH(aq)	5.5–6.5				
Surface area	0.95 m <sup>2</sup> /g				
Porosity	0.76				

### 2.3 Batch Adsorption Studies at Various pH

The effect of pH on the adsorption of Cd(II), Cr(VI) and Pb(II) onto peat was studied by the addition of 1 g peat to 100 ml solution containing 100 mg/l of heavy metal. The pH of the solutions was adjusted to be in the range 2.5–8.0 by the addition of either 0.1 N sulfuric acid solution for samples containing Cd(II) and Cr(VI), and 0.1 N nitric acid solution for samples containing Pb(II), or 0.1 N sodium hydroxide solution.

The suspensions were mixed at 100 rpm for the equilibrium time determined in the kinetic study. At the end of the period, the suspensions were filtered and the concentration of metals was determined by atomic absorption spectroscopy.

### 2.4 Batch Studies at Different Concentration of Heavy Metals

The effect of heavy metal concentration in solution in the range of 10–300 mg/l for Cd(II) and Cr(VI) and in the range of 100–600 mg /l for Pb(II) on the adsorption process was studied by the addition of 1 g peat to 100 ml heavy metal-containing solution. The pH was adjusted to the optimal value for each metal cation and the equilibrium time for each metal was that determined in the kinetic studies. After reaching equilibrium, suspensions were filtered and metals were determined.

## 3 Results and Discussions

### 3.1 Batch Kinetic Studies

The plot of heavy metal concentration in solution versus time, presented in Fig. 1, shows different equilibrium times for the studied metals: 1 h for cadmium(II), in agreement with Gosset et al. (1986);

the equilibrium time for chromium(VI) is high, about 60 h, in agreement with Sharma and Forster (1993), but much higher than that reported by Viraraghavan and Rao (1993). This difference in equilibrium time for Cr(VI) may be due to the difference in concentration of Cr(VI) in aqueous medium in which the batch experiments were conducted: 100 mg Cr(VI) / l in our experiments compared with only 1 mg Cr(VI)/l in the experiment of Viraraghavan and Rao.

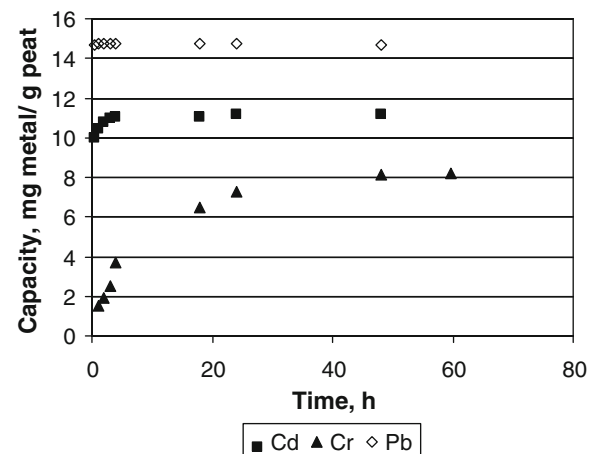
The equilibrium time for lead is 30 min, in agreement with the results reported by Chen et al. (1990) and Couillard (1994); the high affinity of peat for lead being well known.

The batch kinetic data for the adsorption of Cd(II), Cr(VI) and (Pb) onto peat were tested for a first reaction order. The rate equation for a first order reaction is given by Levenspiel 1974:

$$-\ln C_e/C_o = kt,$$

where:

$C_e$  is the concentration of the solute after time  $t$ , milligrams per liter;



**Fig. 1** The influence of contact time on the adsorption capacities of peat towards Cd(II), Cr(VI) and Pb(II) ions

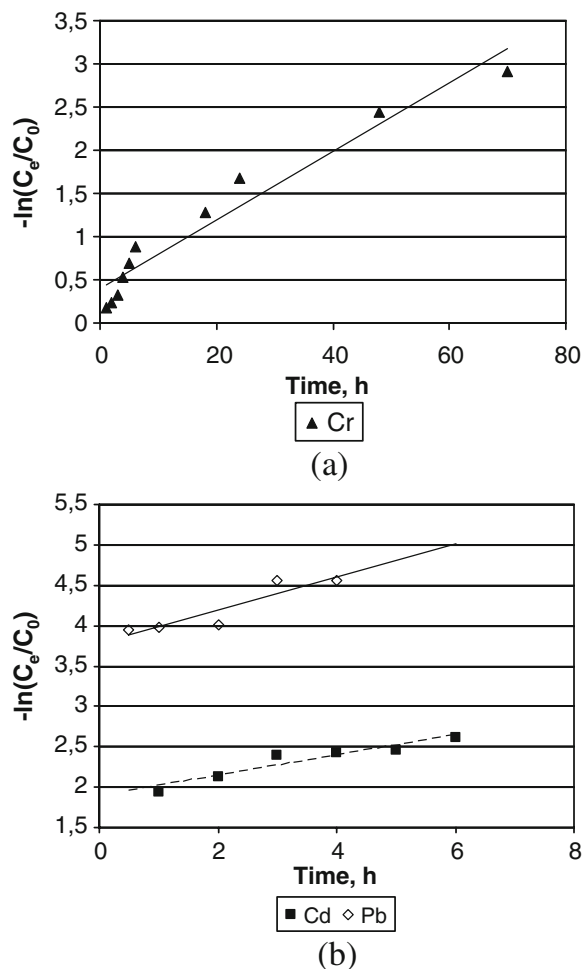
$C_0$  is the initial concentration of solute, milligrams per liter;

$K$  is the reaction rate constant,  $\text{time}^{-1}$ .

The reaction rate constants,  $k$ , for the adsorption of Cd(II), Cr(VI) and Pb(II) onto peat were determined from the plot of  $[\ln(C_e/C_0)]$  versus time, shown in Fig. 2. The values of the reaction rate constants obtained were 0.1250, 0.0396 and 0.2053  $\text{h}^{-1}$  for Cd(II), Cr(VI) and Pb(II), respectively (Table 2). According to these data, the selectivity of peat towards Cd(II), Cr(VI) and Pb(II) in decreasing order is:

Pb(II) > Cd(II) > Cr(VI).

The adsorption of lead and cadmium onto peat is different than the adsorption of chromium, following



**Fig. 2** Linearized plot for reaction rate constants for (a) Cr(VI), (b) Cd(II) and Pb(II)

**Table 2** Reaction rate constants for adsorption of Cd(II), Cr(VI) and Pb(II) onto peat

Metal	Rate constant, $\text{h}^{-1}$ (this research)	Rate constant, $\text{h}^{-1}$ (Viraraghavan and Rao 1993)
Cd(II)	0.1250	0.0134
Cr(VI)	0.0396	0.0379
Pb(II)	0.2053	–

multiple first order kinetics: initial rapid adsorption followed by a very slow adsorption process. The rapid adsorption process occurred in 30 min in the case of lead and in 60 min in the case of cadmium.

To determine whether intraparticle diffusion is the sole rate-limiting mechanism for adsorption, the amount of heavy metal adsorbed versus square root of time is presented in Fig. 3. Weber and Morris (1963) stated that if intraparticle diffusion is the rate-controlling factor, uptake of the adsorbate from solution will vary proportionately with the square root of time. In the plots of Fig. 3, the lines do not pass through the origin; therefore, intraparticle diffusion may not be the sole rate limiting factor (Poots et al. 1976).

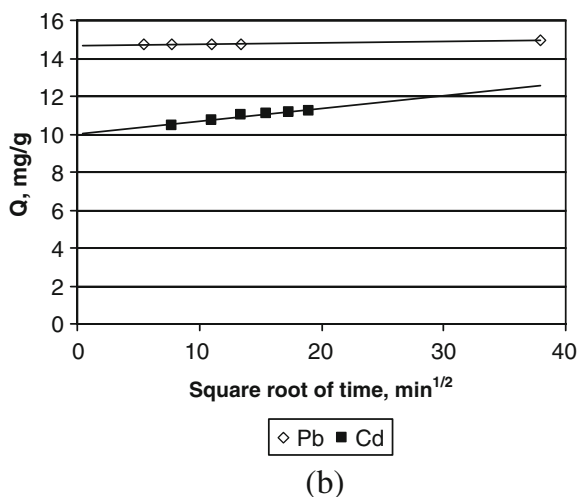
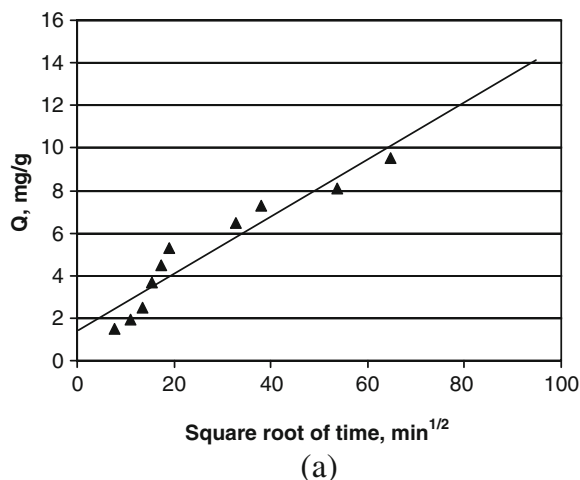
### 3.2 Batch pH Studies

The range of pH studied was chosen to vary between 2 and 6 for Cr(VI) adsorption experiments, between 2.5 and 7.5 for Cd(II) and between 4.5 and 7.0 for Pb(II). These values were chosen to ensure peat stability, as it is well known that peat is not stable below pH 2 and above pH 8, as well as to avoid removal by precipitation of the metal hydroxides.

The effect of pH on the sorption of Cd(II), Cr(VI) and Pb(II) is shown in Fig. 4. The maximum amount of Cd(II) and Cr(VI) adsorbed onto peat occurred at pH 7.5 and 3.5, respectively, whereas no influence of pH was observed for Pb(II) adsorption in the pH range 4.5–7.0.

Figure 5 shows the influence of pH on the adsorption efficiencies of Cd(II), Cr(VI) and Pb(II) onto peat. At pH 7.5, 95% of Cd(II) is removed, at pH 3.5, 94% of Cr(VI) is removed and over 96% of Pb(II) is removed in the pH range 4.5–7.0.

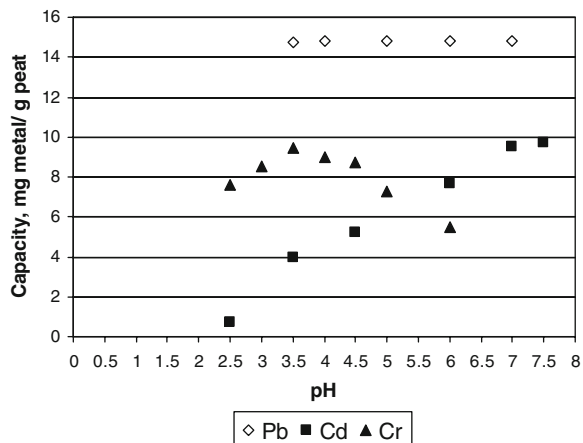
The sorption of these metals onto peat may be explained on the basis of the chemical constituents of peat. The main chemical constituents of peat are humic and fulvic acids, with a high presence of



**Fig. 3** Amount of metals adsorbed on peat versus square root of time (a) Cr(VI), (b) Cd(II) and Pb(II)

anionic groups (Allen 1987). These are compounds with a complex structure; the molecules of humic acids are generally larger than those of fulvic acids and both exhibit an acidic character arising from phenolic and carboxylic groups. Another species present in peat is lignin, a polymeric substance. Although the hydroxyl groups in lignin are less acidic than the phenolic and carboxylic groups of the humic and fulvic acids, they nevertheless exert a repulsive force on the approaching anions. These factors therefore aid the adsorption of heavy metals onto peat.

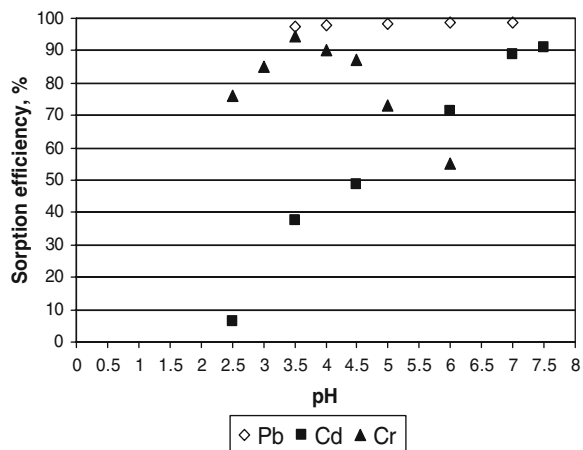
Figures 4 and 5 demonstrate that the effect of pH on ion removal is different for the studied cations, Pb (II), Cd(II), and for the anion Cr(VI). Cd(II) adsorption shows the expected dependence on pH. Higher pH values result in higher metal ion loading, since



**Fig. 4** The influence of solution pH on the adsorption capacities of peat towards Cd(II), Cr(VI) and Pb(II) ions

the dissociation of the low acidic functionalities of the peat become more efficient. At the same time, the metal ion in the solution forms hydroxyl compounds, which have a higher sorption affinity than the cation. Further research is needed for Pb(II) adsorption at higher initial concentrations, since nearly all of the Pb (II) was removed in the range of pH studied.

The pH dependence of Cr(VI) shows a maximum curve. The explanation of this finding needs more detailed consideration of the ion loading mechanism. Peat is not considered a suitable adsorbent for anions, since the carboxyl functionalities using the ion exchange and chelation mechanism are only able to bond cations. A positively charged surface can be formed at very low pH values, which is a likely way for Cr(VI) anion sorption if the matrix of the



**Fig. 5** The influence of pH on removal efficiency of Cd(II), Cr (VI) and Pb(II) onto peat

adsorbent is resistant to oxidation. Bearing in mind the structure and chemical stability of peat, this may be the mechanism for Cr(VI) reduction, whereas the formed Cr(III) will be bound in a similar way to the other tested cations.

The pH dependence found for Cr(VI) removal can be explained as the sum of pH dependence of Cr(VI) reduction and pH dependence of Cr(III) loading. Since the two processes show adverse pH dependence, a maximum shape is expected (Viraraghavan and Rao 1993).

### 3.3 Batch Studies at Different Concentrations of Heavy Metals in Solution

Adsorption isotherms were determined by mixing 1 g of peat with fixed volumes of metal ion solution at different initial concentration, at room temperature.

Bearing in mind the high affinity of peat for lead, the concentration range for this metal was different in comparison with those used for cadmium and chromium. Accordingly, the initial concentration of lead in solution ranged between 150 and 600 mg Pb(II)/l, while the initial concentration of cadmium and chromium ranged between 10 and 300 mg/l.

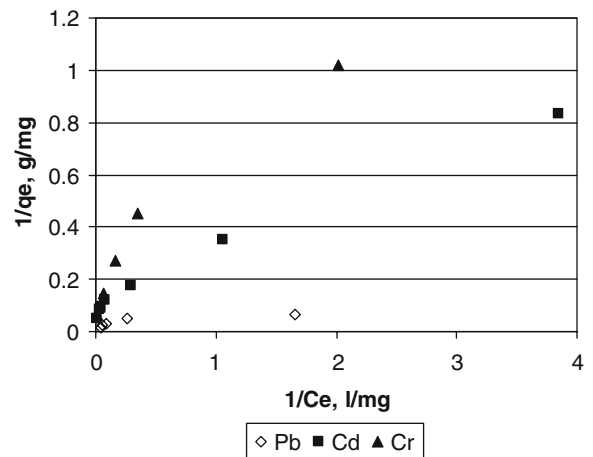
All samples were buffered to the optimum pH previously determined as follows: 3.5 for chromium (VI)-containing solution, 7.0 for cadmium-containing solution and 6.0 for lead-containing solution. The equilibrium times were those previously determined for each metal.

In order to optimize the design of an adsorption system to remove heavy metal ions, it is important to establish the most appropriate correlations for the equilibrium curves for each system (McKay and Porter 1997). In the present study, two isotherm equations were tested, namely those of Langmuir and Freundlich.

The Langmuir isotherm fits the following equation:

$$q_e = \frac{QkC_e}{1 + kC_e}$$

where:  $q_e$  is the metal concentration in the adsorbent,  $k$  is the constant of Langmuir's equation related to the enthalpy of the process,  $Q$  is the adsorption capacity to form the single layer, and  $C_e$  is the metal concentration in the solution. This isotherm is an example of a favorable isotherm and is applicable under the following hypothesis: a uniform surface of the solid, uniform energies of adsorption on the surface, the



**Fig. 6** Langmuir plot for adsorption of Cd(II), Cr(VI) and Pb (II) onto peat

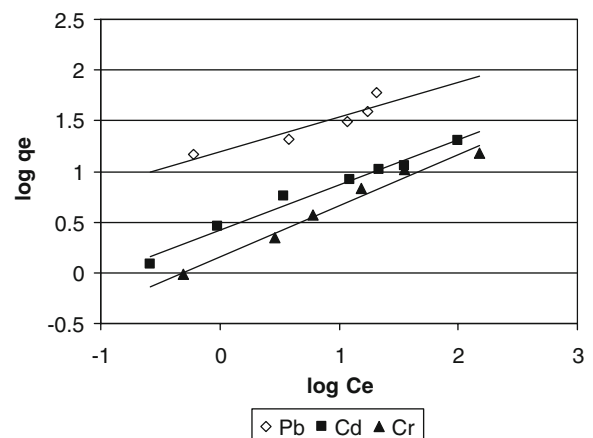
absence of interaction between adsorbed molecules and adsorption in a single layer (no transmigration of sorbate in the plane of the surface).

The  $Q$  parameter gives the theoretical monolayer saturation capacity and is a measure of the amount of metal adsorbed when the monolayer is completed (the Langmuir adsorption isotherm assumed that the adsorbed layer is one molecule in thickness). The Langmuir equation is applicable to homogeneous sorption in which each metal ion-peat sorption process has equal sorption activation energy.

Freundlich's isotherm fits the following equation:

$$q_e = K_F C_e^{1/n}$$

where:  $q_e$  is the amount adsorbed per specified amount of adsorbent, milligrams per gram,  $K_F$  is a constant related to the adsorption capacity,  $C_e$  is the



**Fig. 7** Freundlich plot for adsorption of Cd(II), Cr(VI) and Pb (II) onto peat

**Table 3** Langmuir and Freundlich constants for adsorption of Cd(II), Cr(VI) and Pb(II) onto peat

	Langmuir constants			Freundlich constants		
	$Q$	$k$	Correlation coefficient, $r$	$K_F$	$1/n$	Correlation coefficient, $r$
Lead	36.4960	1.0960	0.8751	15.7943	0.3396	0.9251
Cadmium	10.5708	0.4826	0.9938	2.6479	0.4463	0.9896
Chromium	6.8027	0.3296	0.9704	1.4491	0.5053	0.9897

concentration on the solution, and  $n$  is an empirical parameter related to the intensity of adsorption which varies with the heterogeneity of the adsorbent. For values within the range  $0.1 < 1/n < 1$ , adsorption is favorable (Namasivayam and Yamuna 1992; Raji and Anirudhan 1998).

This model is valid for heterogeneous surfaces and predicts an increase in the concentration of the ionic species adsorbed onto the surface of the solid when increasing the concentration of said species in the liquid phase.

The results of applying the Langmuir and Freundlich adsorption models to the experimental values are shown in Figs. 6 and 7, respectively. The constants derived from these isotherms are presented in Table 3.

The essential characteristics of a Langmuir isotherm can be expressed in terms of a dimensionless constant separation factor or equilibrium parameter, which is defined as (Viraraghavan and Rao 1993):

$$R = 1/[1 + kC_o]$$

where:  $k$  is the Langmuir constant and  $C_o$  is the initial solute concentration, milligrams per liter.

The parameter  $R$  indicates the shape of the isotherm, as follows:

Value of $R$	Type of isotherm
$R > 1$	Unfavorable
$R = 1$	Linear
$0 < R < 1$	Favorable
$R = 0$	Irreversible

Table 4 presents the  $R$  values for the adsorption isotherm of cadmium, chromium and lead onto peat. All the  $R$  values show that cadmium, chromium and lead adsorption onto peat are favorable, being very close to irreversible in the case of lead.

The nature of metal binding onto peat has been extensively researched, although consensus on the exact mechanisms has yet to be reached. All authors agree that the natural capacity of peat to retain cations

is related to the pH of the solution; the optimum pH varies depending on the metal to be removed. A general assumption that can be used for the sorption of most ions onto peat is that the pH should be within the range 3–8 (Brown et al. 2000).

Studying the sorption of Cu(II), Cd(II) and Zn(II) onto peat, McKay and Porter (1997) found that the more electronegative ions were attracted to the peat surface more strongly. Our results regarding Cd(II) and Pb(II) uptake confirm this observation; the electronegativity (according to Pauling) corresponding to these metals is 1.8 and 1.7, respectively. This cannot be applied to Cr(VI), as it is present in solution in anionic form.

#### 4 Conclusions

Horticultural peat is a suitable material for the uptake of Pb(II), Cd(II) and Cr(VI) from aqueous solutions, showing the highest selectivity for Pb(II) and the lowest for Cr(VI).

**Table 4** Equilibrium parameter  $R$  for cadmium, chromium and lead adsorption on horticultural peat

Initial metal concentration, mg/l	Equilibrium parameter ( $R$ )		
	Cadmium	Chromium	Lead
600	–	–	0.0015
425	–	–	0.0020
300	0.0068	0.010	0.0030
150	0.0136	0.021	0.0060
96.5	0.021	–	–
83	–	0.0353	–
60	0.033	–	–
43	–	0.0659	–
30	0.065	–	–
25	–	0.1080	–
12	0.147	–	–
10	–	0.233	–

The kinetic studies indicated that equilibrium time varies as follows: 30 min for Pb(II), 1 h for Cd(II) and 60 h for Cr(VI).

Both Langmuir and Freundlich models achieved good fittings for the three metals, obtaining favorable isotherms.

The adsorption capacity is pH dependent for Cd(II) and Cr(VI), but not for Pb(II); the optimum pH values being 7.5 for Cd(II) and 3.5 for Cr(VI). No dependence of Pb(II) adsorption was observed in the pH range 4.5–7.

Sorption capacities increased with increasing metal concentration in solution. The uptake capacities of peat with respect to these heavy metals (for an initial solution containing 300 mg heavy metal/l) are: 0.288 mmol Cr(VI)/g peat, 0.178 mmol Cd(II)/g peat and 0.145 mmol Pb(II)/g peat.

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