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Biosorption of cadmium (II) and lead (II) from aqueous solutions using mushrooms: A comparative study

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ABSTRACT

Sorption capacity of oyster mushroom (*Pleurotus platypus*), button mushroom (*Agaricus bisporus*) and milky mushroom (*Calocybe indica*) were evaluated on biosorption of heavy metals, viz. cadmium (II) and lead (II) from aqueous solutions. The optimum sorption conditions were studied for each metal separately. The desired pH of the aqueous solution was found to be 6.0 for the removal of cadmium (II) and 5.0 for removal of lead (II) for all the mushrooms. The percent removal of both the metals was found to increase with the increase in biosorbent dosage and contact time. The fitness of the biosorption data for Langmuir and Freundlich adsorption models was investigated. It was found that biosorption of cadmium (II) and lead (II) ions onto the biomass of the three mushrooms were better suitable to Langmuir than Freundlich adsorption model. *P. platypus* showed the highest metal uptake potential for cadmium (q_{max} 34.96 mg/g) whereas *A. bisporus* exhibited maximum potential for lead (q_{max} 33.78 mg/g). Milky mushroom showed the lowest metal uptake capacity for both the metals. The present data confirms that mushrooms may be used as efficient biosorbent for the removal of cadmium (II) and lead (II) ions from aqueous solution.

1. Introduction

Contamination of the aqueous environment by heavy metals due to the discharge of metal containing effluents into the water bodies is one of the most serious environmental issues of the century. The heavy metal pollution represents a significant environmental problem arising from its toxic effects and accumulation throughout the food chain. The removal and recovery of heavy metals from wastewater is significant in the protection of the environment and human health [1].

The conventional methods used for metal removal include chemical precipitation [2], ion-exchange [3], membrane filtration [4] activated carbon adsorption [5], etc. However these treatment methods become less effective and more expensive when situations involving high volumes and low metal concentrations are encountered [6]. The application of membrane processes and activated carbon are also restricted because they are cost intensive. Biosorption technology has gained important credibility during recent years because of its eco-friendly nature, excellent performance, and cost-effectiveness.

Biosorption involves a combination of active and passive transport mechanisms. The first stage, usually referred to as passive uptake, is an initial rapid and reversible accumulation step. The second stage, usually referred to as active uptake, is slower intracellular bioaccumulation, often irreversible and related to metabolic activity [7–10].

Cadmium and lead compounds are being used in a wide variety of commercial processes. Unregulated disposal of cadmium containing effluents in both developing and developed countries has led to the contamination of ground waters. A variety of biomaterials and microorganisms have been explored by researchers for heavy metal biosorption and bioaccumulation including fungi [11], yeast [12], algae [13] and mosses [14] but mushrooms, which belong to the category of macrofungi, are yet to be thoroughly investigated.

Fruit bodies of macrofungi (mushrooms) are considered ideal for the purpose of evaluation as biosorbents, because it has been demonstrated that many fungal species exhibit high biosorptive potentials [8,15–17]. Mushrooms grow prolifically and are found in many parts of the world [18]. They are macro in size, tough in texture and have other physical characteristics conducive for their development as biosorbents without the need for immobilization or deployment of sophisticated reactor configuration as in the case of microorganisms [19]. The aim of the present work was to evaluate the sorption capacity of three different mushrooms, *Pleurotus platypus*, *Agaricus bisporus* and *Calocybe indica* in respect of heavy metals, viz. cadmium and lead from aqueous solutions.

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2. Materials and methods

2.1. Preparation of biosorbent

Fruit bodies of oyster (*P. platypus*) and milky mushrooms (*C. indica*) were procured from the mushroom farm at Krishi Vigyan Kendra of Tamil Nadu Agriculture University at Virinjipuram, Vellore District, Tamil Nadu, India. Fruit bodies of *A. bisporus* used in this study were purchased from a grocery shop at Vellore which was supplied by Saptarishi Agro Industries Ltd., Pazhayanoor, Tamil Nadu, India. Mushrooms were washed thoroughly with deionized water to remove the dirt and impurities and later dried in an oven at 40 °C for 24 h. The dried fruit bodies were then pulverized in a mortar and pestle. Particles with 425–600 μm size were used for the experiment.

2.2. Preparation of cadmium and lead solutions

The aqueous solutions of cadmium (II) and lead (II) used in the present investigation were prepared using water purified by means of a Milli-Q system (Milli-Q® Academic-model, USA). Analytical grade chemicals provided by sd-fine chem. Ltd. were used throughout. Stock solutions of cadmium and lead were prepared by dissolving 2.744 g of $Cd(NO_3)_2 \cdot 4H_2O$ and 1.598 g of $Pb(NO_3)_2$ in 1 l deionized water. The stock solutions were used to prepare dilute solutions of different working concentrations.

2.3. Analysis of metal ions

The concentration of cadmium and lead ions in the biosorption medium was determined using Atomic Absorption Spectrophotometer (Varian AA-240, Australia) equipped with deuterium arc background corrector and air acetylene burner. The hollow cathode lamp was operated at 4 mA for cadmium and 7 mA for lead. Analytical wavelength was set at 228.8 nm for cadmium and 217.0 nm for lead.

2.4. Batch biosorption experiments

The batch biosorption experiments of the three mushrooms were carried out in 250 ml Erlenmeyer flasks containing 0.1 g of the biosorbent in 100 ml of cadmium (II) and lead (II) solutions (10 mg/l) separately at 28 ± 2 °C on a rotary shaker at 120 rpm. The effect of pH on biosorption rate was investigated in a pH range of 2.0–8.0 (which was adjusted with 0.1N HCl or 0.1N NaOH at the beginning of the experiment). The effect of biosorbent dosage was studied by varying the dosage in the range of 0.5–6.0 g/l. For optimization of contact time for biosorption of cadmium (II) and lead (II), desired volumes of metal solutions with a desired dosage of the biosorbent were agitated on an orbital shaker at a desired rpm. Samples were collected at definite time intervals (5, 10, 15, 20, 25, 30, 45, 60, 120, 180, 240, 300, and 360 min), filtered and residual metal concentration in the filtrates were analyzed by Atomic Absorption Spectrophotometer. Batch experiments investigating the effects of initial concentration of cadmium (II) and lead (II) were conducted as above with the metal concentration varied from 10 to 100 mg/l. The metal solutions were filtered after the desired contact time and cadmium and lead ion concentrations in the filtrates were determined.

For each experiment a blank, containing 100 ml of only the cadmium (II) or lead (II) solution without any biosorbent was shaken simultaneously to determine any adsorption of the metal onto the walls of the conical flasks. A control, with 100 ml of deionised water (no metal ion added) and 0.1 g of biosorbent was also shaken to determine any leaching of metal from mushrooms. Each experiment was repeated three times and the results are given as averages.

2.5. Analysis and laboratory treatment of wastewater

The raw wastewater sample was collected from a Common Effluent Treatment Plant (CETP) located in Ranipet, Vellore Dt., Tamil Nadu, India. The physico-chemical characteristics of wastewater sample were analyzed promptly after collection using standard analytical methods [20]. The characteristics of the wastewater were: color: black, pH: 7.5, TDS: 5,534 mg/l, TSS: 2563 mg/l sulfate: 326 mg/l and nitrate 4.4 mg/l. The presence of some heavy metals was also determined using AAS: cadmium: 11.2 mg/l, lead 6.6 mg/l, nickel: 2.3 mg/l, zinc: 3.3 mg/l, copper: 2.4 mg/l. The pH of the wastewater was adjusted to 5.5 using 0.1N HCl. The wastewater was then fed through a glass column (2.5 cm i.d. and 4 cm long) packed with 3.4 g of the biosorbent *P. platypus* at a flow rate of 6.0 ml/min controlled by a peristaltic pump. After treatment, the samples collected from the exit were analyzed for cadmium (II) and lead (II) concentrations by AAS. The wastewater was treated till the values reached to satisfy the EPA standards [21].

3. Results and discussion

It was found from blank and control studies that the adsorption of cadmium and lead onto the walls of the conical flasks was negligible and the mushrooms did not leach any metal into the aqueous solutions.

3.1. Influence of pH

Hydrogen ion concentration in the adsorption is considered as one of the most important parameters that influence the adsorption behaviour of metal ions in aqueous solutions. It affects the solubility of the metal ions in the solution, replaces some of the positive ions found in the active sites and affects the degree of ionization of the adsorbate during the reaction [22]. The effect of initial pH on the biosorption of cadmium (II) and lead (II) ions onto *P. platypus*, *A. bisporus* and *C. indica* were evaluated within the pH range of 2–8. Studies beyond pH 8 were not attempted because precipitation of the ions as hydroxides would be likely [23].

From Figs. 1 and 2, it could be seen that for all the three biosorbents, cadmium and lead adsorption increased along with the increase of pH of the adsorbate solution. This pH dependency of biosorption efficiency could be explained by the functional groups involved in metal uptake and metal chemistry. At low pH values, protons occupy most of the biosorption sites on the biosorbent surface and less cadmium and lead ions could be sorbed because of

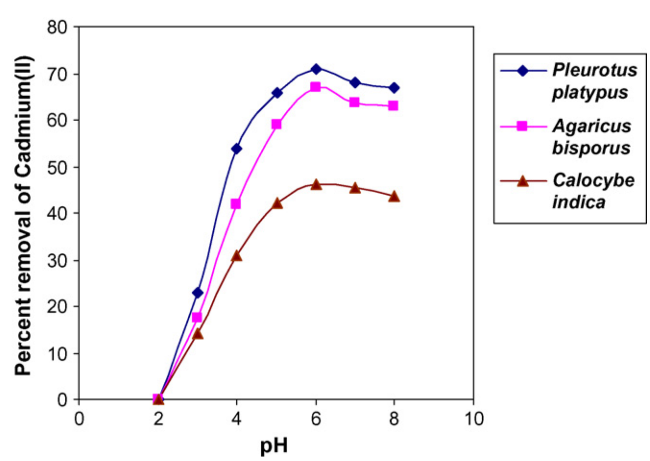

Fig. 1. Effect of pH on cadmium (II) sorption capacity of three different mushrooms (initial cadmium concentration = 10 mg/l, biosorbent dosage = 0.1 g/100 ml, contact time = 6 h).

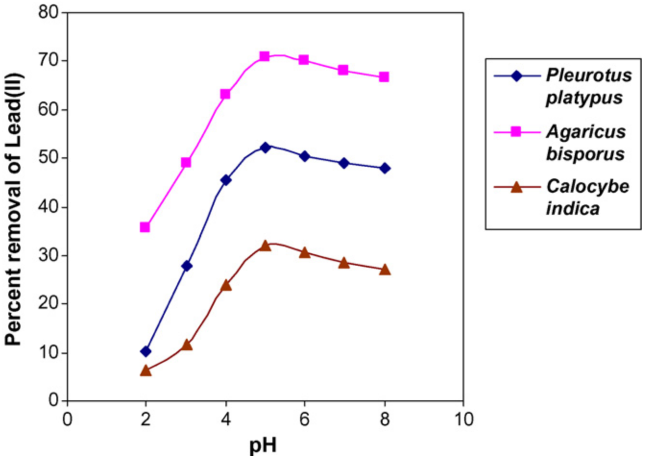

Fig. 2. Effect of pH on lead (II) sorption capacity of three different mushrooms (initial lead concentration = 10 mg/l, biosorbent dosage = 0.1 g/100 ml, contact time = 6 h).

electric repulsion with protons on biosorbent. When the pH values increased, biosorbent surfaces were more negatively charged and the biosorption of metal ions (positive charge) increased and reached equilibrium at pH 6.0 for cadmium and pH 5.0 for lead. Decrease in biosorption at higher pH (pH > 6.0 and >5.0) is due to the formation of soluble hydroxylated complexes of the metal ions and their competition with the active sites, as a consequence, the retention would decrease again.

3.2. Influence of contact time

Biosorption of cadmium (II) and lead (II) by the three biosorbents as a function of time is depicted in Figs. 3 and 4, respectively. In the case of cadmium (II) biosorption by *P. platypus*, the removal efficiency reached equilibrium at 60 min and that by *A. bisporus* and *C. indica* reached equilibrium at 240 min. The respective removal efficiencies of lead by *P. platypus*, *A. bisporus* and *C. indica* reached at 120, 240 and 180 min.

In the initial stages the removal efficiencies of both the metals by all the three biosorbents increased rapidly due to the abundant availability of active binding sites on the biomass, and with gradual occupancy of these sites, the sorption became less efficient in the later stages [24].

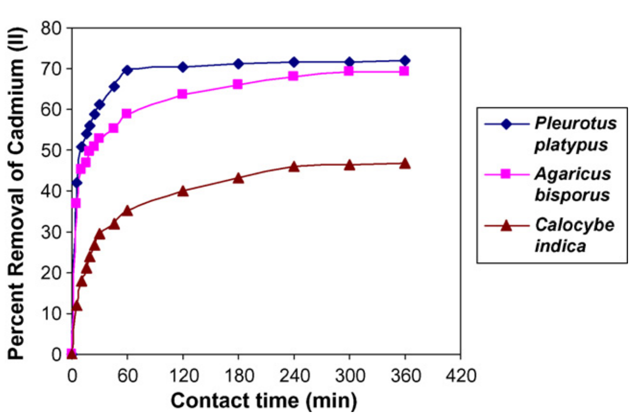

Fig. 3. Effect of contact time on cadmium sorption capacity of three different mushrooms (initial cadmium concentration = 10 mg/l, biosorbent dosage = 0.1 g/100 ml, pH 6.0).

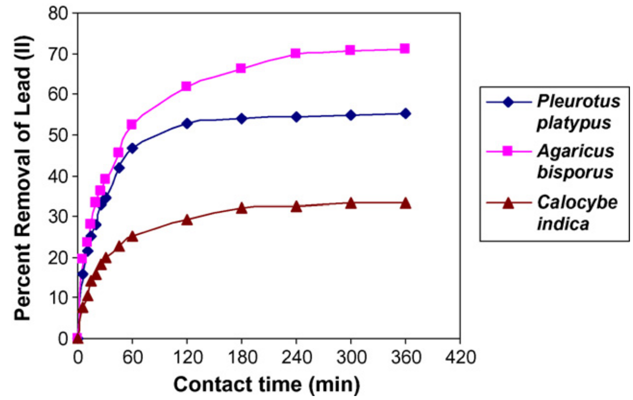

Fig. 4. Effect of contact time on lead (II) sorption capacities of three different mushrooms (initial lead concentration = 10 mg/l, biomass dosage = 0.1 g/l and pH 5.0).

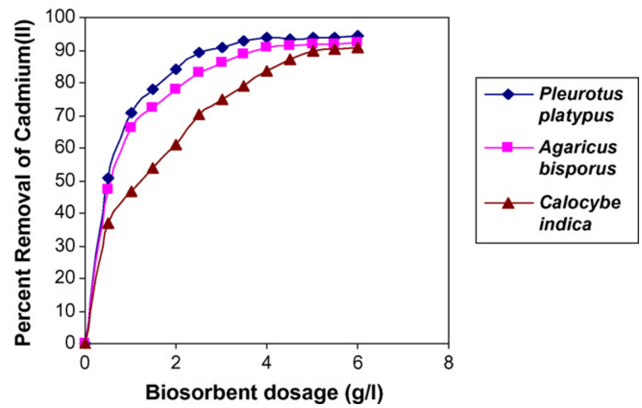

Fig. 5. Effect of biosorbent dosage on cadmium (II) sorption capacity of three different mushrooms (initial cadmium concentration = 10 mg/l, pH 6.0, contact time = 4 h).

3.3. Influence of biomass dosage

The number of available sites and exchanging ions for adsorption depends upon the amount of adsorbent in the biosorption process. The effect of adsorbent concentration on the metal removal efficiency is presented in Figs. 5 and 6. The cadmium uptake was found to increase rapidly with increasing concentration of biosorbent upto a biosorbent dose of 3 g/l in *P. platypus*, 4 g/l in *A. bisporus* and 5 g/l in case of *C. indica*. Similarly in case of lead, the removal efficiency was found to increase with increase in biosorbent dose upto 4 g/l for *P. platypus*, 3 g/l for *A. bisorpus* and 5.5 g/l in case of *C. indica*.

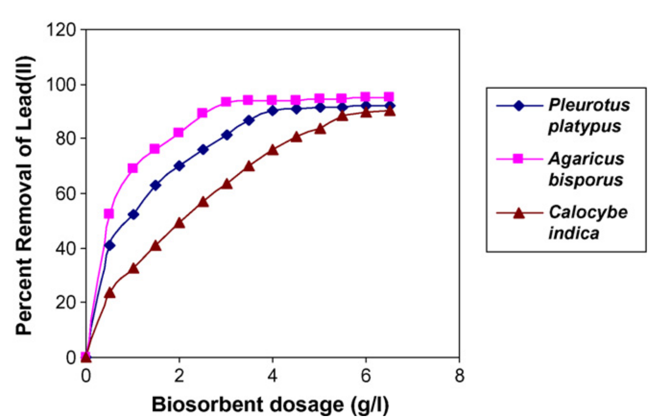

Fig. 6. Effect of biosorbent dosage on lead (II) sorption capacity of three different mushrooms (initial lead concentration = 10 mg/l, pH 5.0, contact time = 4 h).

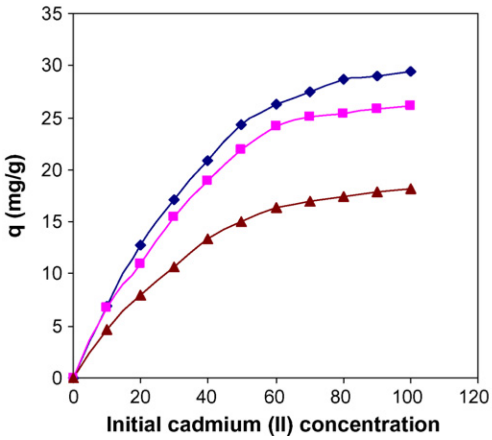

Fig. 7. Effect of initial metal concentration for biosorption of cadmium (II) (initial cadmium concentration = 10–100 mg/l, pH 6.0, contact time = 4 h).

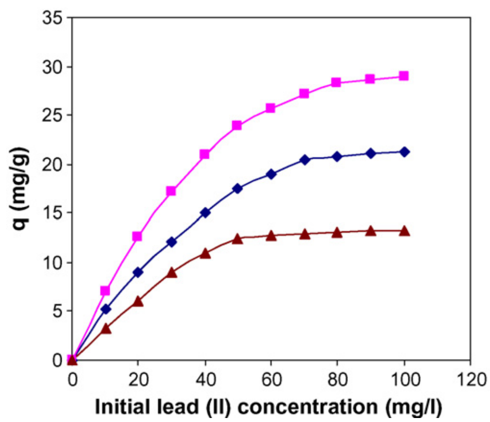

Fig. 8. Effect of initial metal concentration for biosorption of lead (II) (initial lead concentration = 10–100 mg/l, pH 5.0, contact time = 4 h).

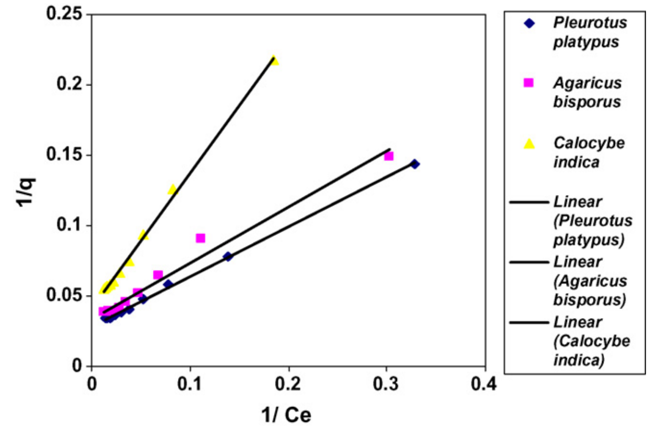

Fig. 9. Langmuir isotherm for biosorption of cadmium (II).

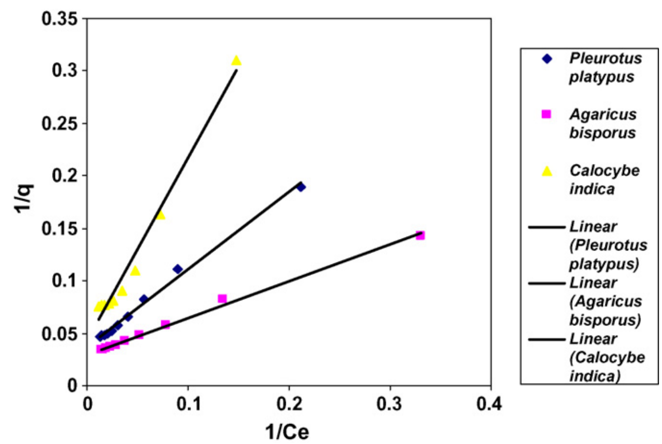

Fig. 10. Langmuir isotherm for biosorption of lead (II).

However, beyond this dosage the increase in removal efficiency was marginal and this may be due to reduction in concentration gradient. The increase in the removal efficiency can be attributed to the increased number of sites and exchangeable ions available for adsorption.

3.4. Influence of initial metal concentration

The initial concentration of the metal (cadmium and lead) in the solution remarkably influenced the equilibrium uptake of cadmium (II) and lead (II), respectively. It was noted that initial concentration increased the sorption of cadmium (II) and lead (II) as is generally expected due to equilibrium process (Figs. 7 and 8). This increase in uptake capacity of the three biosorbents with the increase in initial metal concentrations is due to higher availability of metal ions (cadmium and lead), for the sorption. Moreover, higher initial concentration provides increased driving force to overcome all mass transfer resistance of metal ions between the aqueous and solid phase resulting in higher probability of collision between metal ions and sorbents. This also results in higher metal uptake [25].

3.5. Biosorption isotherm models

Many sorption isotherm models are usually used to fit the adsorption data in order to obtain a linear regression data to predict the maximum adsorption capacity of the adsorbent. Langmuir and Freundlich models are the most widely used models in the case of the adsorption of metal ions with biosorbents. Langmuir model suggests monolayer sorption on a homogeneous surface without interaction between sorbed molecules. In addition, the model assumes uniform energies of sorption onto the surface and no transmigration of the sorbate.

In view of the values of linear regression coefficients (R^2) given in Tables 1 and 2, it was noted that the Langmuir isotherm model exhibited better fit to the sorption data of both cadmium (II) and lead (II) for all the three mushrooms than the Freundlich isotherm model. Figs. 9 and 10 indicate the linear relationship between the amount (mg) of cadmium (II) and lead (II) ions sorbed per unit

Table 1
Isotherm parameters for cadmium (II) sorption (initial cadmium (II) concentration = 10–100 mg/l, pH 6.0, biosorbent dosage 1.0 g/l, temperature 28 ± 2 °C).

S. No.	Mushrooms	Langmuir parameters			Freundlich parameters		
		q_{max} (mg/g)	b (l/mg)	R^2	n	K_f	R^2
1.	*Pleurotus platypus*	34.96	0.0809	0.9991	2.2217	4.9980	0.9488
2.	*Agaricus bisporus*	29.67	0.0850	0.9791	2.2079	4.3092	0.9523
3.	*Calocybe indica*	24.09	0.0432	0.9972	1.9984	2.2750	0.9498

Table 2
Isotherm parameters for lead (II) sorption (initial lead (II) concentration = 10–100 mg/l, pH 5.0, biosorbent dosage 1.0 g/l, temperature 28 ± 2 °C).

S. No.	Mushrooms	Langmuir parameters			Freundlich parameters		
		q_{max} (mg/g)	b (l/mg)	R^2	n	K_f	R^2
1.	*Pleurotus platypus*	27.10	0.0499	0.9937	1.9996	2.7580	0.9669
2.	*Agaricus bisporus*	33.78	0.0846	0.9961	2.2301	4.9522	0.9509
3.	*Calocybe indica*	23.41	0.0244	0.9816	1.8796	1.5017	0.8780

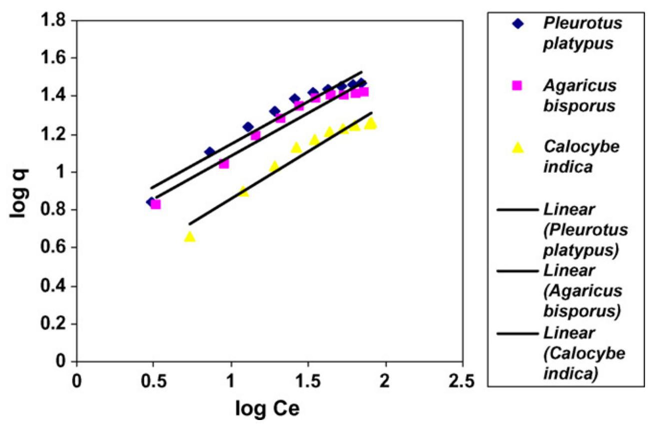

Fig. 11. Freundlich isotherm for biosorption of cadmium (II).

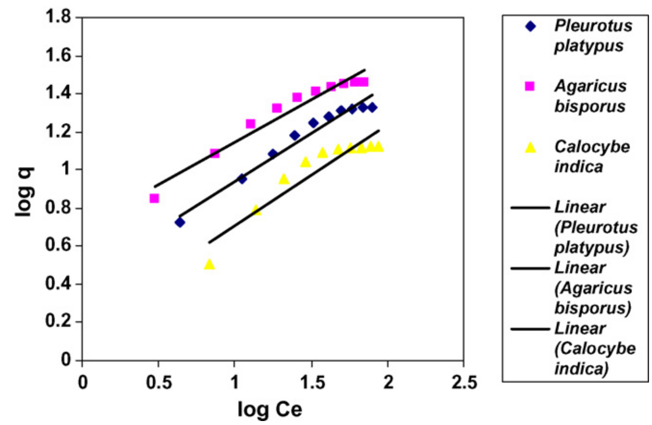

Fig. 12. Freundlich isotherm for biosorption of lead (II).

mass of *P. platypus*, *A. bisporus* and *C. indica* against the concentration of cadmium (II) and lead (II) ions remaining in solution (mg/l). Figs. 11 and 12 show the Freundlich isotherms obtained for biosorption of cadmium (II) and lead (II) ions onto the three different mushrooms. The R^2 values shown in Tables 1 and 2 indicated that Freundlich model was not able to adequately describe the relationship between the amounts of cadmium (II) and lead (II) adsorbed by the biomass and its equilibrium concentration in the solution. However, the Langmuir model best fitted the equilibrium data since it presents higher R^2 values.

Model parameters obtained for three different mushrooms for cadmium and lead biosorption are given in Tables 1 and 2. It can be noteworthy that *P. platypus* is more potential biosorbent for cadmium (II) sorption and *A. bisporus* for lead (II) whereas low biosorption capacity was noted in case of *C. indica* for both the metals.

The cost of the adsorbent is an important issue that must be considered while selecting an adsorbent. The cost of commercial activated carbon varies from \$5 to \$6/kg (approximately) [26], whereas the cost of cultivated mushrooms chosen for the present

Table 3
Cadmium (II) adsorption on mushrooms in comparison with other adsorbents.

Biosorbent	Experimental conditions		Cadmiun adsorption capacity (mg/g)	References
	pH	Metal concentration (mg/l)		
Pleurotus platypus	6.0	10–100	34.96	Present study
Agaricus bisporus	6.0	10–100	29.67	Present study
Calocybe indica	6.0	10–100	24.09	Present study
Phanerocheate chrysosporium	6.0	5–500	27.79	[27]
Papaya wood	5.0	5–500	17.22	[28]
Aspergillus niger	–	10	3.74	[29]
Saccharomyces cerevisae	–	0.5–50	31.20	[30]
Saccharomyces cerevisiae	7.2	8–30	8.17	[31]
Mucor roxii	5.0	–	8.36	[32]
Nile water algae	6.0	4–44	29.86	[33]
Ceramium virgatum	5.0	10–400	39.70	[34]

Table 4
Lead (II) adsorption on mushrooms in comparison with other adsorbents.

	Experimental conditions		Lead adsorption capacity (mg/g)	References
	pH	Lead concentration (mg/l)		
Pleurotus platypus	5.0	10–100	27.10	Present study
Agaricus bisporus	5.0	10–100	33.78	Present study
Calocybe indica	5.0	10–100	23.41	Present study
Pseudomonas putida	4.5	0–1000	36.0	[35]
Penicillium sp.	6.4	50	6.1	[36]
Cephalosporium aphidicola	5.0	0–10	36.9	[37]
Rhizopus arrhizus	5.0	0–300	15.5	[38]
Mucor rouxii	5.0	–	25.22	[32]
Citrobacter freundii	4.0	0–481	42.5	[39]

study is less than $3 on a retail basis. Based on the results obtained with the use of other biosorbents in the removal of cadmium and lead reported by other workers (Tables 3 and 4), it can be concluded that mushrooms as cost-effective biosorbents may play an important role for removing heavy metals from aqueous environment.

In order to demonstrate the practical application of operating conditions during biosorption process in the present study, column experiments were conducted with wastewater adjusting the pH to 5.5 and the efficiency of the process was checked.

Choice of *P. platypus* was found to be more economical for field application considering its low cost of cultivation compared to *A. bisporus*. Thus further work was carried out to confirm the suitability of *P. platypus* to remove metals from real wastewater in a column mode. The results showed that about 3.4 g of biosorbent was sufficient to treat a wastewater volume of 5.2 l containing the metals (Cd (II) and Pb (II)). The wastewater was treated till the metal concentrations reached the values established by EPA which is 0.5 mg/l and 1.3 mg/l for cadmium and lead, respectively. Therefore, the present study confirmed that mushrooms could be used for the treatment of wastewater containing cadmium and lead.

4. Conclusion

The effectiveness of the mushrooms as biosorbent of heavy metals was confirmed. The effect of parameters like pH, biomass dosage, contact time and initial metal ion concentration during the biosorption process was noted. The desired pH value for biosorption was found to be 6.0 for cadmium and 5.0 for lead, respectively for the mushrooms under study. Higher biosorbent dosages resulted in higher percent removal of cadmium (II) and lead (II) due to abundant availability of sorption sites and exchanging ions. The desired biosorbent dosage in case of *P. platypus* for cadmium (II) and lead (II) sorption was 3 and 4 g/l, respectively whereas in case of *A. bisporus*, it was 4 and 3 g/l, respectively. Langmuir isotherm model exhibited better fit to the sorption data of both cadmium (II) and lead (II) for all the three mushrooms than the Freundlich isotherm model. Higher values of q_{max} (34.96 mg/g) and K_f (4.9980) in case of *P. platypus* for cadmium sorption and q_{max} (33.78 mg/g) and K_f (4.9522) for lead sorption of *A. bisporus* indicate their greater suitability for removal for cadmium (II) and lead (II) ions, respectively from wastewater. But *C. indica* exhibited lower potential for biosorption of both cadmium (II) and lead (II) ions. This difference in uptake capacities may be due to the differences in the cell wall composition of the mushrooms.

Based on the results of column studies conducted with real wastewater, it may be concluded that the mushrooms could be used as potential biosorbents for the removal of cadmium (II) and lead (II) from aqueous solution. Further research is in progress to elucidate the mechanism involved during the biosorption process.

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