

## Removal of Some Heavy Metals from Medicinal Plants (*Indigofera tinctoria* & *Acalypha Indica*) Using Activated Carbon as Adsorbents

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### ABSTRACT

Heavy metals may be harmful to humans through ingestion of medicinal plants containing metals through atmospheric pollution, pesticides and chemical fertilizers root uptake, ingestion of plants splashed with contaminated soil. Metals of concern are arsenic (As), cadmium (Cd), copper (Cu), Mercury (Hg) and lead (Pb). Removal of the heavy metal ions from contaminated medicinal plants *Indigofera tinctoria* and *Acalypha indica* by adsorption technique. It is the best, economically viable and efficient method. It, therefore, becomes necessary to remove these heavy metals from medicinal plants by an appropriate treatment before consuming.

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### Introduction

Arsenic is one of the contaminants found in the environment which is notoriously toxic to man and other living organisms. Cadmium is an irritant to the respiratory tract and prolonged exposure to this pollutant can cause anemia and a yellow stain that gradually appears on the joints of the teeth (Sharma 2008). Exposure of copper has resulted in jaundice and enlarged liver. It is suspected to be responsible for one form of metal fume fever (Bobsa 2007). Mercury causes impairment of pulmonary function and kidney, chest pain and dyspnoea. Lead poisoning causes damage to liver, kidney and reduction in hemoglobin formation, mental retardation, infertility and abnormalities in pregnancy (Sas-Nowosielska et al 2008). The equilibrium of lead, copper, gold, iron, mercury, silver, tin and zinc are seen in ayurvedic and as essential for normal functioning of human body and an important component of good healthy in addition, some products contain other heavy metals such as arsenic (Huff et al 2007). One needs to first ask the question if the medicines were herbal. If the products reported were herbal, then one needs to know the basal level of heavy metal accumulation by the herbs. Therefore the objective of this work was to investigate heavy metal content in selected Indian medicinal plants (Mahwash Zahra Kirmani et al 2011).

### Materials and Methods

#### Instrumentation

Atomic adsorption spectrometer (AAS) 6300 (Shimadzu, Japan) operating with an air acetylene flame was used to analyze the concentration of heavy metals. All measurements were repeated three times and those results in which the standard deviations were found. The pH measurements were performed with a controlled pH analyzer (Systronics). All the adsorption experiments carried out for agitating the sample for a desired contact time.

### Chemicals

Analytical grade reagents were used for heavy metal solution, ACS reagent grade concentrated nitric acid, NaOH and pH buffer solutions (E. Merck) were used to adjust pH values of samples. In all experimental work, distilled demineralized water was used.

#### Medicinal plants solution

The medicinal plants are washed, dried in shade and ground into a fine powder (< 0.5 mm) using an analytical laboratory grinder. A weighed amount of plant material is placed in a crucible and ashed by heating in a muffle furnace gradually through different stages. This ensures total destruction of organic matter. The residual ash is then treated with either nitric acid or hydrochloric acid and dried, the process is repeated, till a clear solution is obtained. The dried residue is taken in 3% nitric acid warmed filtered and made up to a known volume (Djeridane et al 2006).

#### Adsorbent

Granulated activated carbon used as adsorbent, the percent heavy metal removal was calculated using the following equation

$$\text{Metal ions removal (\%)} = (C_0 - C_e) \times 100 / C_0 \quad (1)$$

Where  $C_0$ : initial metal ion concentration of test solution, mg/l;  $C_e$ : final equilibrium concentration of test solution,

#### Treatment techniques

The adsorption of heavy metals on Granularity Activated Carbon (GAC) was studied by batch technique. A known weight of adsorbent Granular activated carbon (2.0 g adsorbent) was stirred with 250 ml of medicinal plant solution containing heavy metals, it is evident from the results that the contact time required to attain equilibrium is dependent on the initial concentration of heavy metals (Vaclavikova 2008). The optimal contact time to attain equilibrium with activated carbon was experimentally found to be about (7-8 hrs.).

## Results and Discussion

### Adsorption studies of heavy metals

The parameters which warrant the evaluation for investigating the mechanism of adsorption and deciding the efficiency of the simultaneous removal of the metal ions by adsorption on GAC is Concentration of the metal ions , Dose of the adsorbents, Contact time between adsorbate and adsorbent, pH of the solution, Stirring speed.

#### Effect of Initial Concentration of the metal ions

Adsorption studies of heavy metal As (III), Cd(II), Cu(II), Hg(II) and Pb(II) ions on GAC at a fixed dose of adsorbent (2g/250ml for GAC) at different initial concentrations of the metal ions As ranged from 0.048-1.147 µg/g, cadmium 0.05-2.02 µg/g, copper 10.08-53.62 µg/g, mercury 0.009-0.158 µg/g and lead 13.85 -27.97 µg/g. The contact time (1-7 hrs) and pH 6.5- 7.1 for GAC, at temperature 28± 1°C were carried out. The variations in the percentage removal of metals ion with its concentration are shown in relevant data obtained from these experiments are given in table 1. In GAC adsorbents, it was observed that the percentage of removal of heavy metals ions is low at higher concentrations and gradually increases the concentration of the heavy metals ions are decreases (Neogi et al 2008). This is due to the fact that after the formation of mono-ions layer at concentration over the adsorbent surfaces, further formation of the layer is highly hindered at higher concentration due to the interaction between metal ions present on the surface and in the solution. In addition to that, at low concentration of the heavy metal ions, the rates of the initial number of moles of heavy metals ions to the available surface area of the adsorbent is large and subsequently, the formation of the adsorption becomes independent of the initial concentration of the metal ion. But at higher concentrations, the adsorption sites available for the adsorption become lesser, and hence the percentage removal of the metal at higher concentration decreases (WHO 2018).

#### Effect of contact time

Contact time is another effective factor in batch adsorption technique. In order to study the effect of contact time on the removal of the As (III), Cd(II), Cu(II), Hg(II) and Pb(II) ions, experiments were conducted at different contact times from 1-7 hrs for GAC keeping the optimum concentrations and dosage, pH 6-6.5 and temperature 28 ± 1°

C. The variation of the percentage removal of the As (III), Cd(II), Cu(II), Hg(II) and Pb(II) ions by adsorption of GAC with contact time is shown in table 1. The extent of removal of the heavy metal ions increases initially and then stagnant after the optimum contact time. For GAC the optimum contact time was 7 hrs for the removal of 96.77 % 95.23%,93.25,94.0 and 93.0% ) As (III), Cd(II), Cu(II), Hg(II) and Pb(II) ions (Kumar 1995).

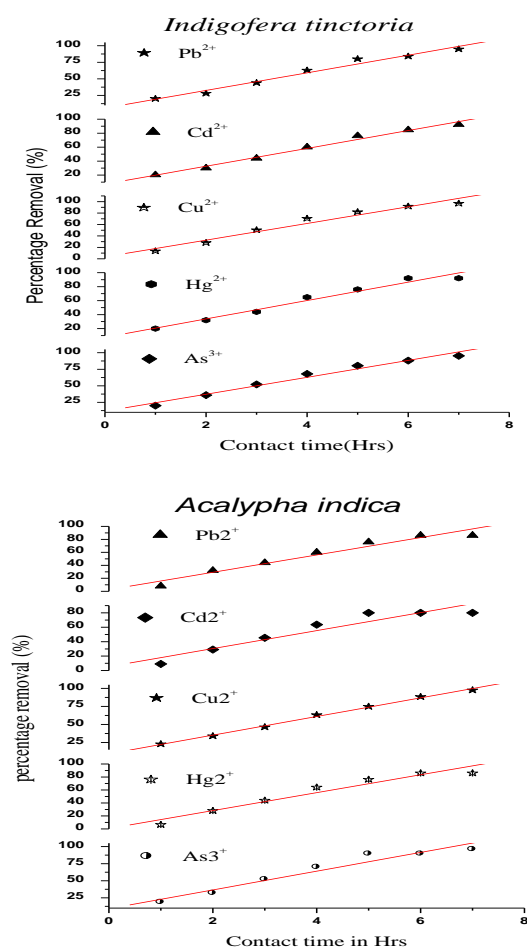
#### Kinetics of adsorption

In order to study the kinetics of adsorption of the metal ions, the batch type adsorption experiments were carried out by varying the contact time at optimum initial concentration of the metal ions and fixed dose of adsorbents (2.0/250 g/ml of GAC) at 28 ± 1° C. The boundary layer resistance will affect the rate of adsorption. Increase in contact time will reduce this resistance and increase the mobility of the adsorbate (heavy metal ions) in the adsorption system. The mechanism of removal of heavy metal ions by adsorption on the GAC and NPAC from medicinal plants may be assumed to involve four stages viz., a) Migration of the heavy metal ions from the bulk of the solution to the surface of the adsorbent ,b) Diffusion through the boundary layer to the surface of the adsorbent, c) Adsorption at active sites ,d) Intra-particle diffusion of the ions into the interior pores of the adsorbent particles(Vaclavikova 2008 ).

The applicability of various first order kinetic equations like Natarajan-khalaf, Lagergren, Elovich and powder functions equations were tested. The values of rate constant from the above rate equations are collected in table 1. The linear kinetic plot Figures 1&2 observed and computed (correlation coefficient) values which are very close to unity indicate the applicability of these first order kinetic equations and the first order nature of the adsorption process of the metal ions. The rate of adsorption of heavy metal ions such as As (III), Cd(II), Cu(II), Hg(II) and Pb(II) on GAC is good as expected level. Apart from the adsorption at the outer surface of the adsorbent, there was also a possibility was explored by plotting the amount of As (III), Cd(II), Cu(II), Hg(II) and Pb(II) ions adsorbed (x/m) per unit mass of the adsorbent against time <sup>1/2</sup>. The linear plots was observed and shown in Figure 1&2, indicates that the intra-particle diffusion was the rate limiting step (Vaclavikova 2008).

**Table-1. Effect of contact time percentage removal of heavy metal ions by adsorption on granulated activated carbon**

Medicinal plants, part used Time (hrs)	Time (hrs)	Quantity of Heavy metals adsorbed (ppm)				
		As (Initial Conc 0.234)	Cd (Initial Conc (1.43)	Cu (Initial Conc 15.21)	Hg (Initial Conc 0.043)	Pb (Initial Conc 69.94)
<i>Indigofera tinctoria</i> (whole plant)	1	0.187 (20.09)	1.14 (20.28)	13.16 (13.48)	0.034 (20.00)	55.95 (20.00)
	2	0.150 (35.90)	1.00 (30.07)	10.95 (28.01)	0.029 (32.09)	50.35 (28.01)
	3	0.112 (52.14)	0.80 (44.06)	7.55 (50.36)	0.024 (43.95)	39.16 (44.01)
	4	0.075 (67.95)	0.57 (60.14)	4.47 (70.61)	0.015 (64.88)	25.97 (62.87)
	5	0.046 (80.34)	0.34 (76.22)	2.75 (81.92)	0.010 (76.05)	13.98 (80.01)
	6	0.028 (88.03)	0.22 (84.62)	1.25 (91.78)	0.003 (92.09)	11.19 (84.00)
	7	0.009 (96.15)	0.11 (92.31)	0.50 (96.71)	0.003 (92.09)	3.59 (94.87)
<i>Acalypha indica</i> (whole plant)		Initial ( 0.765)	Initial ( 0.55)	Initial (31.24)	Initial ( 0.046)	Initial ( 48.59)
	1	0.621 (18.82)	0.50 (9.09)	24.10 (22.86)	0.043 (6.56)	44.70 (8.01)
	2	0.520 (32.03)	0.39 (29.09)	20.65 (33.90)	0.033 (28.01)	33.04 (32.00)
	3	0.367 (52.03)	0.30 (45.45)	16.75 (46.38)	0.026 (43.98)	27.21 (44.00)
	4	0.229 (70.07)	0.20 (63.64)	11.50 (63.19)	0.016 (64.11)	19.43 (60.01)
	5	0.122 (84.05)	0.11 (80.00)	7.85 (74.87)	0.011 (76.15)	11.66 (76.00)
	6	0.061 (92.03)	0.08 (84.45)	3.65 (88.32)	0.006 (85.97)	6.78 (86.05)
7	0.030 (96.08)	0.02 (96.36)	0.75 (97.60)	0.006 (85.97)	6.78 (86.05)	



**Figure 1 & 2 Effect of contact time % removal of heavy metal ions by adsorption on granulated activated carbon.**

#### Conclusion

Granulated activated Carbon showed nearly 100% adsorptive removal of heavy metal ions under optimized conditions. These experimental on adsorbents would be quite useful in developing an appropriate technology for the removal of heavy metal ions from contaminated medicinal plants.

#### Reference

Abebe D, Debella A, Urga K. (2003) Medicinal Plants and Other Useful Plants of Ethiopia. Camerapix Publishers International, Singapore, 54-61.  
 Bbosa GS, Kyegombe DB, Ogwal-Okeng J, Bukenya-Ziraba R, Odyek O, Waako P (2007) Antibacterial activity of Mangifera indica (L.), African Journal of Ecology 45: 13-16

Djeridane, A., Yousfi, M., Nadjemi, B., Boutassouna, D., Stocker, P. & Vidal, N. (2006), Antioxidant activity of some Algerian medicinal plants Extracts containing phenolic compounds, Food Chemistry, 97, 654–660

GSB, Pohlit AM (2008) Screening of Amazonian plants from the adolpho ducke forest reserve, Manaus, state of Amazonas, Brazil, for antimicrobial activity. Mem Inst Oswaldo Cruz, Rio de Janeiro 103: 31-38.

Kumar, P. Dushenkov, V. Motto, H. and Rashkin, I., Phytoextraction, (1995) The use of plants to remove heavy metals from soils. Environ Sci. and Technology. 29: 1232-1238

Huff J, Lunn RM, Waalkes MP, Tomatis L, Infante PF(2007), Cadmiuminduced cancers in animals and in humans, International Journal of Occupational and Environmental Health; 13: 202-212.

Mahwash Zahra Kirmani, Sheikh Mohiuddin, (2011), Determination of some toxic and essential trace metals in some medicinal and edible plants of Karachi city, Journal of Basic and Applied Sciences Vol. 7, No. 2, 89-95, (2011).

Mohan.D and C. U. Pittman Jr., (2007), Arsenic removal from water/wastewater using Adsorbents-a critical review, Journal of Hazardous Materials, (2007) vol. 142, no. 1-2, pp. 1–53,.

Neogi U, Saumya R, Mishra RK, Raju KC (2008) Lipid content and in vitro antimicrobial activity of oil of some Indian medicinal plants, Current Research in Bacteriology 1: 1-6.

Sas-Nowosielska A., Galimska-Stypa R., Kucharski R., Zielonka U., Małkowski E., and L. Gray(2008), Remediation aspect of microbial changes of plant rhizosphere in mercury contaminated soil, Environmental Monitoring and Assessment, 137, 1–3,101–109.

Sharma A, Shanker C, Tyagi L, Singh M, Rao C.V.(2008), Herbal Medicine for Market Potential in India: An Overview, Academic Journal of Plant Sciences 1, 26-36.

Vaclavikova.M, Gallios, G. P. Hredzak. S., and S. Jakabsky,(2008),Removal of arsenic from water streams: an overview of available techniques, Clean Technologies and Environmental Policy, 10( 1), 89–95.

WHO (2018) Environmental Health Criteria 101, Methyl Mercury, Geneva, World Health Organization 78.