

Efficiency of Removal of Lead, Cadmium, Copper and Zinc from Aqueous Solutions Using Six Common Types of Plant Leaves

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Abstract: Dry plant leaves of thyme, sage, banana, mint, anise and oleander plants have been used for removing lead, cadmium, copper and zinc ions for aqueous solutions. The most efficient types of plant leaves for the removal process have been found to be oleander leaves for removing lead and copper and sage leaves for removing cadmium and zinc. The maximum efficiency of removal of the studied metal ions by plant leaves was equal to that obtained by activated carbon. The concentration of metal ion in solution has been found to have only slight effect on the efficiency of the removal process. Oven-dried leaves have a little higher efficiency of removal of metal ions than naturally dried leaves.

Key Words: Efficiency, Removal, Thyme, Sage, Banana, Mint, Oleander, Anise, Plant Leaves, Lead, Cadmium, Copper, Zinc

Introduction

Pollution of water with metal ions has been a matter of great concern for the past several years. The scarcity of water sources in several areas of the world, especially in the Middle East, does not allow losing any of these sources through pollution. Thus, finding ways for removing pollution is of prime interest.

Several processes have been proposed for the removal of toxic metal ions from polluted water. These include precipitation (Scollt, 1977), ion exchange (Greig, 1996), electrodeposition (Robinson and Rhodes, 1980) and filtration (Gulson *et al.*, 1997).

Common sorbents such as activated carbon (Ferro-Garcia *et al.*, 1988), fly-ash (Yadava *et al.*, 1987) and zeolite (Kesraouiouki *et al.*, 1994) are extensively used for removing toxic metals from polluted water. The most commonly used sorbent being activated carbon that requires re-generation and is generally costly.

Low cost biological-originated and agriculture by-products and aquatic sorbents have been recommended as cheap sorbents for the removal of toxic metals. Examples of these sorbents are hop (Torresdey *et al.*, 2002), peat (Gosset *et al.*, 1987), moss (Wilhelm *et al.*, 1989), hair (Singh *et al.*, 1993) tea-leaves (Chaudhari and Tare, 1996) and several types of aquatic plants (Ornes, 1994; Lee and Hardy, 1987; Sobhan and Sternberg, 1999 and Aderhold *et al.*, 1996).

Plant leaves are natural wastes that have been found efficient in removing considerable amounts of metal ions from polluted water (Salim and Robinson, 1985; Salim, 1988; Salim *et al.*, 1992; Salim *et al.*, 1994; Sayrafi *et al.*, 1996 and Al-Subu *et al.*, 2001). Examples of proposed plant leaves as efficient in removing metal ions from water are reed (Salim and Robinson, 1985) for cadmium, poplar for lead and copper (Salim and Robinson, 1985 and Salim *et al.*, 1992), cinchona for copper (Salim *et al.*, 1992), pine for nickel (Salim *et al.*, 1994) and cypress for aluminum (Sayrafi *et al.*, 1996).

In the present work, we aim to try some new types of plant leaves which are commonly planted in our area (Palestine and Jordan) for removing lead, cadmium, copper and zinc from aqueous solutions. The efficiency of this removal process is compared with the efficiency of removal using activated carbon.

Materials and Methods

Dry leaves were gathered from the campus of Al-al-Bayt University. Leaves were cleaned thoroughly with de-ionized water and then dried at room temperature. After dryness, leaves were ground, sieved into constant size (40-50 mesh) and then kept in plastic bags ready for use. All leaves used were found free from lead, cadmium, copper and zinc as indicated from the negligible concentrations of these metals leached from leaf-suspensions.

Removal of elements was measured by following the decrease in the concentration of metal ion in aqueous leaf suspensions without adjustment of pH. The concentration of leaves used was always 5 g /100mL. All ranges of metal ion concentrations used in this work were within the linear range of calibration graphs of these metals by the AAS method used in this work. The loss of concentration of metal ions from solution was followed until after steady values. These steady values were generally obtained after about three to four days but readings were followed for about a week.

The instrument used for the AAS measurements was Unicam 929 equipped with a Laminar Flow Burner. Acetylene and air used were of high-pure quality.

A small tube with a sintered glass disk was immersed in the bottle containing leaf suspension to allow having a small amount of clear solution inside the bottle used for the analysis of elements by FAAS. Each experiment was done in triplicate and the average of the three readings was taken. The concentration of element was determined by comparison with calibration graphs. A QC sample, in the same concentration range of measured samples, was analyzed with each group of readings to check the accuracy of readings. All experiments were done at room temperature (20 °C). Old and frequently used polyethylene bottles (250mL) were used in this work. This type of containers was found to cause the minimum loss of metal ions on container surfaces (Salim, 1980). These were decontaminated by soaking in 1M HNO₃ for three days and then washed thoroughly with distilled water. Adsorption of lead, cadmium and copper on container surfaces was found to be negligible on containers used in this work. In addition, all measurements were done against a blank of de-ionized water.

The metal solutions used in this study were prepared by dilution from ready standard stock solutions (1000 mg/L) supplied by the manufacturers of the ASS instrument. Other chemicals used were all of high-grade quality.

Oven dried sage leaves were obtained by drying plant leaves at 90 C for ~ 1 hour in air-circulated oven.

Results and Discussion

Effect of Type of Leaves: Removal of lead from 2.7 mg/L lead solution was studied on the six types of leaves used in this work. The efficiency of these leaves on removing lead ions was compared with activated carbon. The results obtained are shown on Fig. 1. These results show that the maximum efficiency of removal of lead by plant leaves varied between 72-99 % of lead. The efficiency of removal of lead is arranged in the decreasing order:

Activated carbon ~ oleander > mint > thyme > sage > banana > anise.

Cadmium ions were removed from 2.2 mg/L cadmium solutions using the six types of leaves in addition to activated carbon. The results (Fig. 2) indicate a maximum efficiency of removal ranging between 64 - 99 % of the cadmium ions in solution. The efficiency of removal is arranged in the decreasing order:

Activated carbon > sage > oleander > thyme > banana > mint > anise.

Copper ions were removed from 3.2 mg/L copper solutions using the above types of leaves plus carbon. The results obtained are shown on Fig. 3. These results show that the maximum efficiency of removal of copper using plant leaves varied between 47-99%.

The efficiency of removal of the copper is arranged in the decreasing order:

Activated carbon > oleander > thyme > mint > sage > banana > anise.

Zinc ions were removed from 2.7 mg/L zinc solutions using the above six types of leaves in addition to activated carbon. The results (Fig. 4) showed a maximum efficiency of removal by plant leaves ranging between 37 - 99 % of zinc from solution. The efficiency of removal of zinc by the various types of plant leaves, in addition to carbon, can be arranged in the following decreasing order:

Activated carbon ~ sage > oleander > thyme > banana > mint > anise.

Comparing the maximum efficiency of removal of the above metal ions with the efficiency of removal of other types of leaves studied previously for removal of metal ions suggests a better maximum efficiency of removal obtained in this work than before. Examples of previously reported maximum efficiency of removal of metal ions by plant leaves were 88% of Cu on poplar leaves (Al-Subu *et al.*, 2001), 96% of Cd⁺² on reed leaves (Sayrafi *et al.*, 1996), ~ 40% of Cd⁺² on beech leaves (Salim *et al.*, 1992), ~ 80% of Pb⁺² on poplar leaves (Salim *et al.*, 1994), ~ 82% of Zn⁺² on poplar and walnut leaves (Salim, 1980).

Effect of Metal Ion Concentration: Various concentrations of lead, cadmium, copper and zinc ions were used for removal experiments using 50 g/L dry thyme leaves. The results obtained are shown in Table 1. These results show only very little effect of concentration of metal ions on its removal from solution.

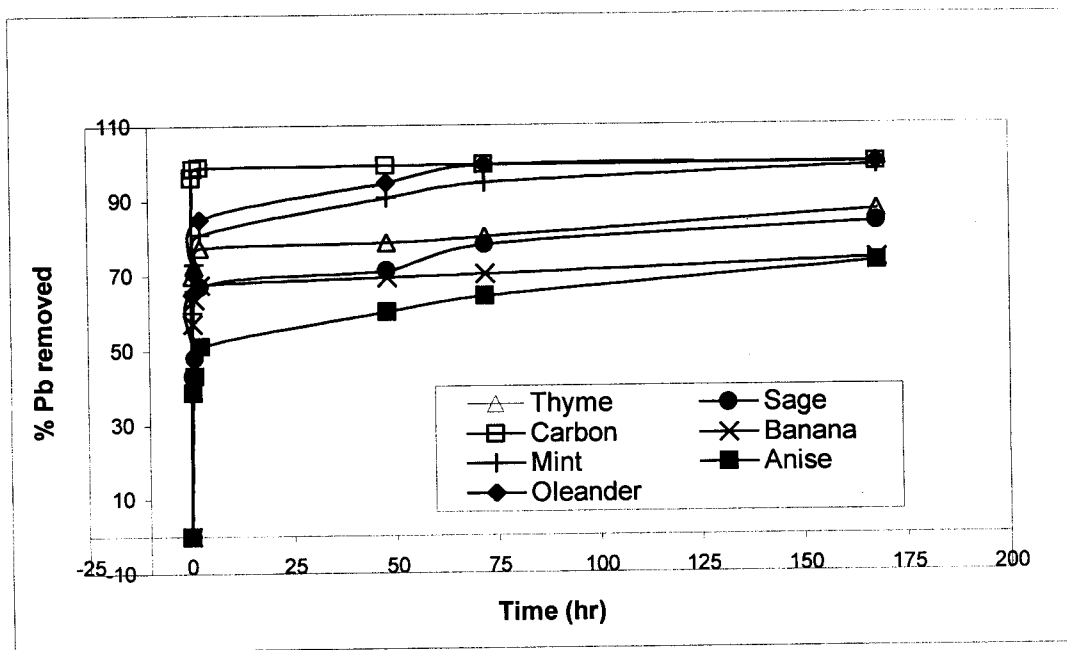


Fig.1: Removal of Lead Ions from 2.7 mg/L Aqueous Lead Solutions using 50 g/L Naturally-Dried Plant Leaves

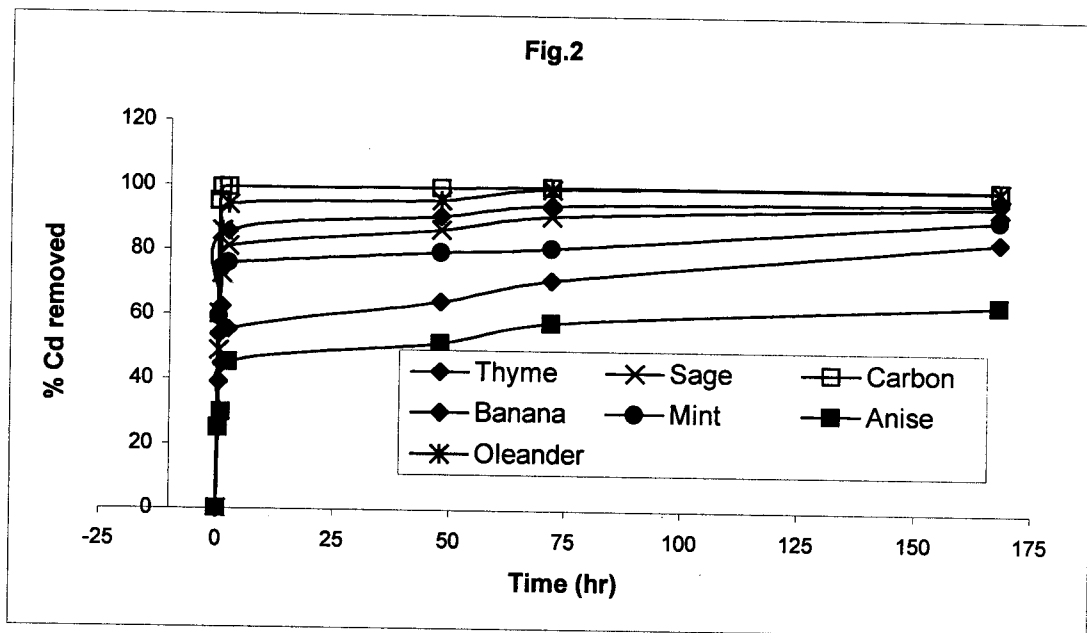


Fig.2: Removal of Cadmium Ions from 2.2 mg/L Aqueous Cadmium Solutions Using 50g/L Naturally Dried Plant Leaves

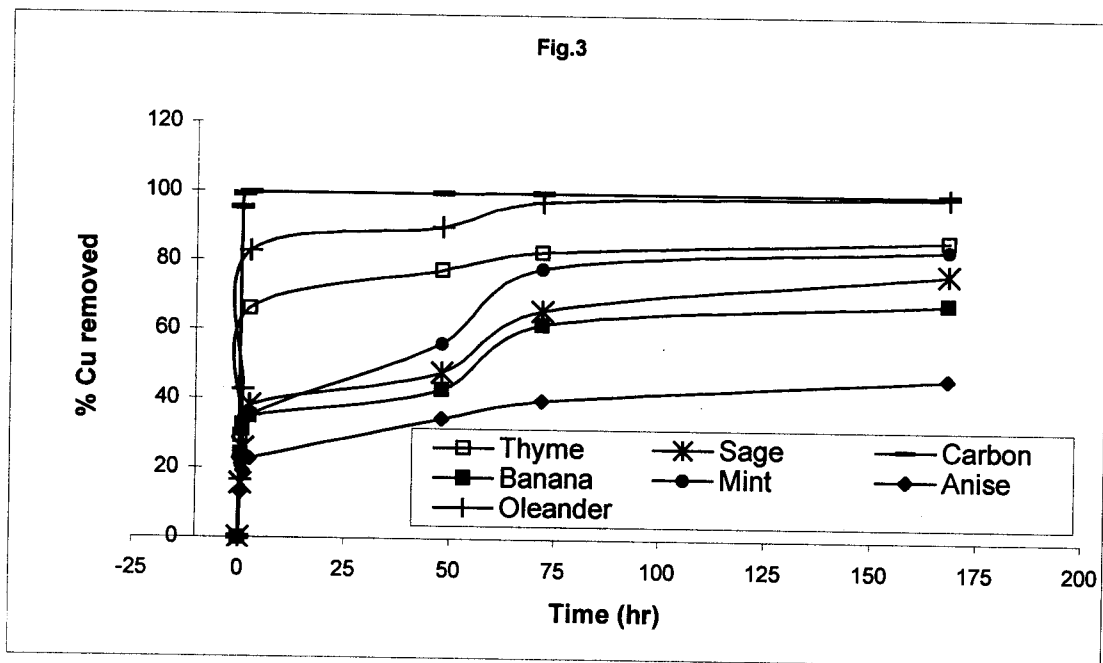


Fig.3: Removal of Copper Ions from 3.2 mg/L Aqueous Cadmium Solutions Using 50g/L Naturally Dried Plant Leaves

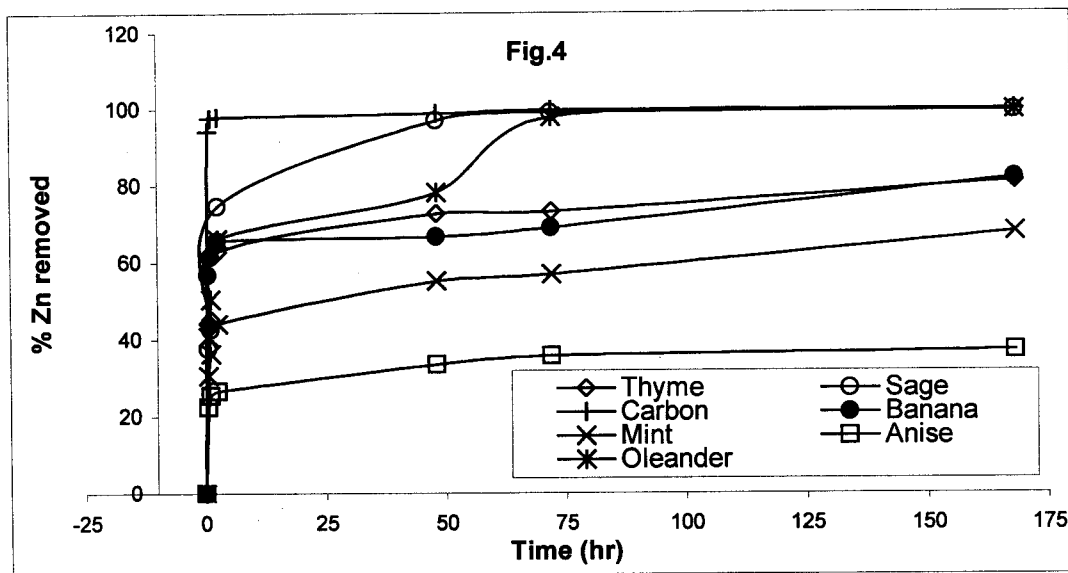


Fig.4: Removal of Zinc Ions from 2.7 mg/L Aqueous Cadmium Solutions Using 50g/L Naturally Dried Plant Leaves

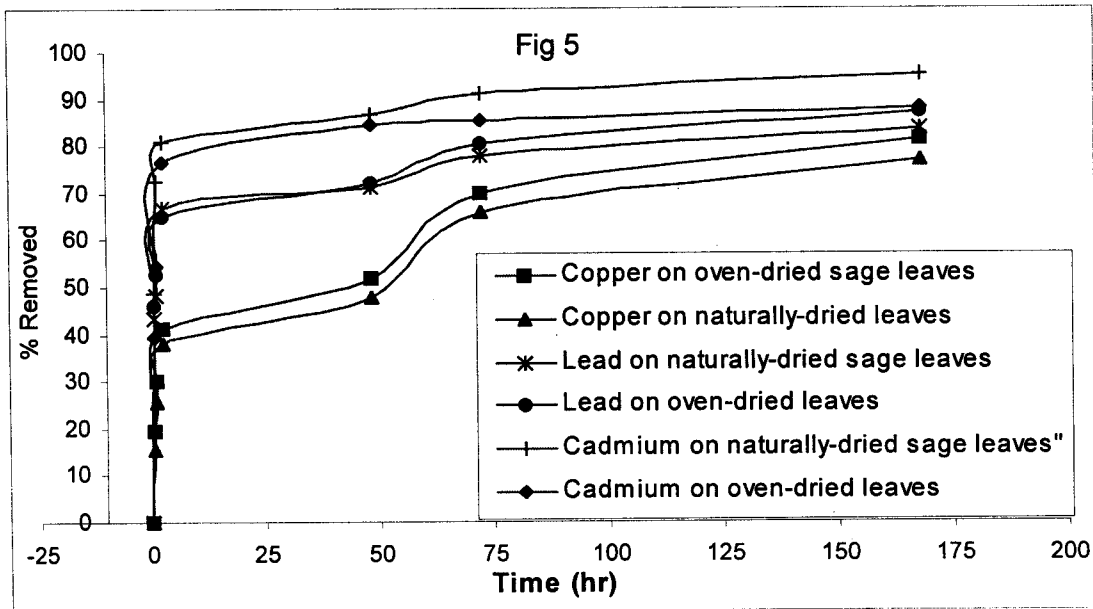


Fig.5: Removal of Lead, Cadmium and Copper from 3.2, 2.4 and 2.7 g/L Metal Ion Solution, Respectively, Using Naturally-Dried and Oven-Dried 50 g/L Sage Leaves

Table 1: Removal of Lead, Cadmium, Copper and Zinc Ions from Various Concentrations of these Metals Using 50 g/L Naturally Dried Thyme Leaves

Time (hr)	Lead					
	Conc. (mg/L)	% Removed	Conc. (mg/L)	% Removed	Conc. (mg/L)	% Removed
0.0	1.37	0.00	3.20	0.00	8.02	0.00
0.5	0.62	54.57	0.96	69.92	2.10	73.81
1.0	0.52	62.11	0.87	72.67	1.92	76.10
2.5	0.35	74.25	0.72	77.46	1.42	82.28
48.0	0.32	76.52	0.68	78.67	1.21	84.88
72.0	0.29	78.79	0.64	80.11	1.08	86.53
168.0	0.19	85.96	0.41	87.12	0.92	88.51
Time (hr)	Cadmium					
	Conc. (mg/L)	% Removed	Conc. (mg/L)	% Removed	Conc. (mg/L)	% Removed
0.0	2.38	0.00	4.57	0.00	7.52	0.00
0.5	1.10	53.82	2.21	51.65	3.10	58.76
1.0	0.90	62.25	0.87	81.03	1.69	77.57
2.5	0.34	85.57	0.65	85.89	1.17	84.50
48.0	0.22	90.73	0.52	88.60	0.92	87.75
72.0	0.13	94.38	0.44	90.46	0.72	90.41
168.0	0.10	95.93	0.21	95.36	0.32	95.73
Time (hr)	Copper					
	Conc. (mg/L)	% Removed	Conc. (mg/L)	% Removed	Conc. (mg/L)	% Removed
0.0	2.74	0.00	4.83	0.00	8.91	0.00
0.5	1.64	29.21	3.23	33.12	6.52	26.79
1.0	1.86	32.31	3.14	35.07	6.21	30.33
2.5	0.93	65.97	1.47	69.55	3.63	59.27
48.0	0.62	77.35	1.32	72.66	3.12	64.96
72.0	0.53	80.60	1.26	73.92	2.89	76.53
168.0	0.42	84.65	0.80	83.40	1.92	78.44
Time (hr)	Zinc					
	Conc. (mg/L)	%Removed	Conc. (mg/L)	% Removed	Conc. (mg/L)	% Removed
0.0	2.74	0.00	4.63	0.00	6.88	0.00
0.5	1.53	44.17	3.12	32.52	5.31	22.80
1.0	1.50	45.33	2.96	63.11	5.18	24.78
2.5	1.02	62.80	1.84	60.32	3.03	56.01
48.0	0.75	72.61	0.81	82.44	1.62	76.44
72.0	0.73	73.23	0.78	83.20	1.46	78.84
168.0	0.52	81.00	0.62	86.68	0.98	85.74

Effect of Drying Leaves: Naturally dried sage leaves were compared with oven-dried leaves for their removal of copper ions from 2.7 mg/L copper solutions, lead ions from 3.2 mg/L lead solutions and cadmium ions from 2.4 mg/L cadmium solutions. The results (Fig. 5) indicate only a little effect of the method of drying leaves with oven-dried leaves being a little more efficient for the removal process than the naturally dried leaves. This agrees with previous results on the effect of dryness of removal of lead, cadmium and copper on other types of plant leaves (Salim *et al.*, 1994; Salim, 2002 and Al-Subu *et al.*, 2001).

Conclusion

In this work six common types of plant leaves were studied for removing lead, cadmium, copper and zinc from aqueous solutions. Plant leaves were generally capable of removing metal ions from solution. The efficiency of the removal process depended highly on the type of leaves used. The most efficient types of leaves were oleander leaves for removing lead and copper ions and sage leaves for removing cadmium and zinc ions from their aqueous solutions. The maximum efficiency of removal of metal ions using the tested plant leaves was equal to that obtained by using activated carbon. This maximum efficiency of removal

was higher than efficiencies previously reported for removing the same metal ions on other types of plant leaves.

Metal ion concentration did not show effect on the efficiency of the removal process of these metals on thyme leaves.

The method of drying leaves showed only a little effect on the efficiency of the removal process with oven-dried leaves being a little more efficient on removing lead, cadmium and copper from their aqueous solutions.

References

Aderhold, D., C. J. Williams and R. G. J. Edyvean, 1996. The removal of heavy-metal ions by seaweeds and their derivatives. *Bioresour. Technol.* 58, 1-6.

Al-Subu, M., R. Salim; I. Abu-Shqair and K. Swaileh, 2001. Removal of dissolved copper from polluted water using plant leaves: Effects of acidity and plant species. *Rev. Int. Contam. Ambient.* 17, 91-96.

Al-Subu, M. M., R. Salim; H. Braik and K. M. Swailehi 2001. Removal of dissolved copper from polluted water using plant leaves: Effects of copper concentration, plant leaves, competing ions and other factors. *Rev. Int. Contam. Ambient.* 17, 123-127.

- Chaudhari, S. and V. Tare, 1996. Analysis and evaluation of heavy metal uptake and release by insoluble starch-xanthate in aqueous environment.
- Ferro-Garcia, M. A., J. R. Utrilla; J. R. Gordillo and I. B. Toledo, 1988. Adsorption of zinc, cadmium and copper on activated carbon obtained from agricultural by-products. *Carbon* 26, 363-373.
- Greig, I. A., 1996. Ion exchange developments and applications. Royal Soc. Chem., Cambridge, U.K.
- Gulson, B.L., A. Sheehan and A.M. Giblin, 1997. The efficiency of removal of lead and other elements from domestic drinking waters using a bench-top water filter. *Sys. Sci. Tot. Environ.* 3, 205-216.
- Gosset, T., J. L. Trancart and D. R. Thevenot, 1986. Batch metal removal by peat. Kinetics and thermodynamics. *Wat. Res.* 20, 21-26.
- Kesraouiouki, S., C. R. Cheeseman and R. Perry, 1994. Natural zeolite utilization in pollution control - a review of application to metal effluents. *J. Chem. Tech. Biotech.* 59, 121-126.
- Lee, T. A. and J. K. Hardy, 1987. Copper uptake by water hyacinth. *Ibid.* A22, 141-160.
- Ornes, W. H., 1994. Bioaccumulation of cadmium in duckweed. *J. Environ. Sci. Health A* 29, 10 35-1044.
- Robinson, J. W. and I. Rhodes, 1980. Dev. of an electrochemical technique for removal of ultra - trace levels of heavy metal from water using accelerated electro-deposition. *Spec.Lett.* 13, 69-92.
- Singh, D. K., D. P. Tiwari and D. N. Saksena 1993. Removal of lead from aqueous solutions by chemically treated tea leaves. *Indian J. Environ. Health* 35,169-177
- Sobhan, R. and S. Sternberg, 1999. Cadmium removal using *Cldophora*. *Ibid* A34, 53-72.
- Salim, R. and J. W. Robinson, 1985. Removal of dissolved aluminum released by acid rain using decaying leaves. *J. Environ. Sci. Health* A20, 701-734.
- Salim, R., 1988. Removal of nickel (II) from polluted water using decaying leaves. *Ibid.* A23, 183-197 and 321-334.
- Salim, R., M. Al-Subu and E. Sahrhage, 1992. Uptake of cadmium from water by beech leaves. *Ibid.* A27, 603-627.
- Salim, R., M. Al-Subu and S. Qashoa, 1994. Removal of lead from polluted water using decaying plant leaves. *Ibid.* A29, 2087-2114.
- Sayrafi, O., R. Salim and S. Sayrafi, 1996. Removal of cadmium from polluted water using decaying leaves. *Ibid.* A31, 2503-2513.
- Salim, R., 1980. Adsorption of lead on container surfaces. *J. Electroanal. Chem.* 106, 251-262.
- Salim, R., M. Al-subu; I. Abu-Shqair and H. Braik, 2003. Removal of zinc from aqueous solutions by dry plant leaves. *Proc. Safety.Environ. Prot., in course of publication.*
- Salim, R. and R. Abu-El-Halawa, 2002. Efficiency of dry plant leaves (mulch) for removal of lead, cadmium and copper from aqueous solutions. *Proc. Safety. Environ. Prot.,* 80,270-276.
- Scolt, M.S., 1977. Sulfex; A new process Tech. for removal of heavy metals from water streams. *Ann Arbor Sci., Michigan.*
- Torresdey, J. G., M. Hejazi; K. Tiemann; J. G. Parsons; M. D. Gardea and J. Henning, 2002. Use of hop (*Humulus lupulus*) Agri. by-products for the reduction of aqueous lead (II) environmental health hazards. *J. Hazard. Mat.* 91, 95-112.
- Wilhelm, M., F. K. Ohnesorge; I. Lombeck and D. Hafner, 1989. Uptake of aluminum, cadmium, copper, lead and zinc from aqueous solutions by human scalp hair and elution of the adsorbed metals. *J. Anal. Toxicol.* 13, 17-21.
- Yadava, K. P., K. K. Panday; B. S. Tyaji and V. N. Singh, 1987. Fly ash for the treatment of Cd (II) rich effluents. *J. Env. Tech.Let.* 8. 225-234.