IChemE
www.ingentaselect.com/titles/09575820.htm

0957–5820/03/$23.50+0.00

Trans IChemE, Vol 81, Part B, July 2003

REMOVAL OF ZINC FROM AQUEOUS SOLUTIONS BY DRY PLANT LEAVES

R. SALIM, M. AL-SUBU, I. ABU-SHQAIR and H. BRAIK

Chemistry Department, An-Najah University, Nablus, Palestine

Removal of zinc from aqueous solutions by 15 species of plant leaves was studied. The maximum efficiency of removal was found to be by walnut and poplar leaves at pH 6 with a maximum removal of 82%. Increasing the concentration of plant leaves increased the removal of zinc up to a limit. Agitation of solution increased the efficiency of the removal process. The presence of competing and complexing agents affected the removal process negatively but also positively in few cases.

Keywords: zinc; removal; plant-leaves; aqueous solutions.

INTRODUCTION

The presence of heavy metals in the environment is a major concern due to their toxicity to man and animal and plant life. Some heavy metals such as lead, cadmium and mercury are considered toxic even at low concentrations. Other heavy metals such as zinc and copper are considered essential at low concentrations but become toxic at high concentration levels. Zinc, in small concentrations, is an essential element for living organisms. It is essential for the enzymes required for forming red blood cells in living organisms. It is also essential for plants because it takes part in the biosynthesis of nucleus acids and polypeptides required for plants. On the other hand, when concentration of zinc increases above a limit (e.g. the threshold limit value (TLV) for zinc in drinking water is $3\,\mathrm{mg\,l^{-1}}$), it becomes toxic to man, animals and plant life (Brooks, 1978; Moore and Ramamoorthy, 1984; Dojlido and Best, 1993). Toxicity of zinc becomes more severe when present with other heavy metals, such as cadmium, in water because it has synergistic effect with these metals. The main sources of zinc to the environment are mining operations, secondary metalproduction, coal combustion, rubber tire wear and phosphate fertilizers (Brooks, 1978; Moore and Ramamoorthy, 1984; Dojlido and Best, 1993; Hammond and Beliles, 1980).

Removal of heavy metals from water and wastewater is very important in order to protect public health. Several methods have been suggested for removing heavy metals from polluted water. These include precipitation, coagulation, complexation, ozonation, ion exchange, electroplating and electrodialysis. Common sorbents such as activated carbon, fly ash, ferrites, zeolite and aluminum oxide have been also used for the removal process.

Low-cost biosorbents are attracting attention as simple and cheap methods for removal of toxic heavy metals from polluted water. Examples of these biosorbents are hair (Wilhelm *et al.*, 1989), wool (Kobayashi and Nishi, 1974); tealeaf (Singh *et al.*, 1993), apple-residues (Lee *et al.*, 1998), seaweed (Aderhold *et al.*, 1996), algae (Gin *et al.*, 2002), and moss (Al-Asheh and Duvnjak, 1997) and aquatic plants (Ornes, 1994; Lee and Hardy, 1987; Sobhan and Sternberg, 1999).

Plant leaves have been suggested as natural, simple and cheap sorbents for efficient removal of several metal ions from polluted water (Salim, 1988; Salim *et al.*, 1992, 1994; Al-Subu *et al.*, 2001; Sayrafi *et al.*, 1999; Salim and Robinson, 1985; Salim and Abu-El-Halawa, 2002). Several factors have been found to affect the removal of heavy metal by plant leaves; these include type of leaves, acidity, concentration of leaves, concentration of metal ions, presence of competing ions and method of treatment of leaves.

The efficiency of plant leaves, at optimal conditions, was proved to be high for several heavy metal ions. The most efficient plant leaves for removing nickel from polluted water was pine leaves at neutral pH values (Salim, 1988). Poplar leaves were found best for removing lead (Salim *et al.*, 1994) and copper (Al-Subu *et al.*, 2001) while reed leaves were best for removing cadmium (Sayrafi *et al.*, 1999) from aqueous solutions. The aim of this paper is to attempt a large number of common plant leaves in Palestine to remove zinc ions from aqueous solutions. The optimal conditions for increasing the efficiency of this removal process will also be investigated.

Finding efficient types of plant leaves for removal of zinc ions might be of interest for industrial and for environmental applications for the removal of zinc from industrial effluents and from polluted water without much additional cost.

METHODS AND MATERIALS

Naturally dried leaves of 15 different plant species common in Palestine were collected from Nablus and Jenin areas in North West Bank (a map is shown in Figure 1). A list of these leaves and their collection sites is shown in Table 1. Leaves were cleaned thoroughly with de-ionized water, dried

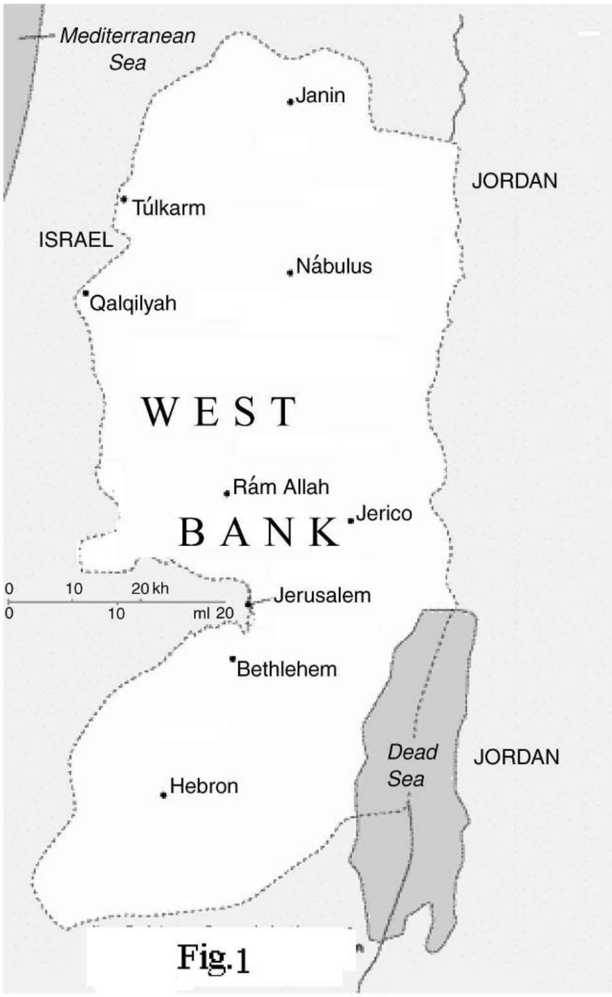

Figure 1. Map of the West Bank, Palestine.

at room temperature, ground, sieved into constant size (40–50 mesh) and finally kept in plastic bags for further use in the experiments. All leaves employed were found free from zinc, as indicated from the negligible concentration of leached zinc ions in solutions of these leaves when soaked in 0.1 M HNO_3 for 2 days.

Table 1. Plant species studied and their collections.

English name	Scientific name	Collection site
Carob	*Ceratonia siliqua* L.	Sieleh Harthieh/Jenin
Cinchona	*Eucalyptus longifolia* L.	Sieleh Harthieh/Jenin
Cypress	*Cupressus sempervirens* L.	Nablus
Ficus	*Ficus nitida* L.	Nablus
Guava	*Psidium guajava* L.	Sieleh Harthieh/Jenin
Loquat	*Eriobotrya japonica* L.	Sieleh Harthieh/Jenin
Olive	*Olea europea* L.	Sieleh Harthieh/Jenin
Oak	*Quercus ithaburensis* L.	Sieleh Harthieh/Jenin
Pine	*Pinus halepensis* L.	Sieleh Harthieh/Jenin
Pistache	*Pistacia palaestina* L.	Sieleh Harthieh/Jenin
Poplar	*Populus euphratica* L.	El-Badan/Nablus
Reed	*Phragmites australis* L.	Till/Nablus
Sisso	*Dalbergia sisso* L.	Nablus
Walnut	*Juglans regia* L.	Nablus
Willow	*Salix alba* L.	El-Badan/Nablus

Frequently used glass bottles (250 ml) were employed in this work for the batch sorption tests. These bottles were decontaminated by soaking in 1 M HNO_3 for 3 days and then they were washed thoroughly with distilled water. Adsorption of zinc by the container surfaces was found to be negligible under the conditions of the present work (Salim and Hilal, 1986). In addition, all measurements were done against a blank of deionized water.

Zinc solutions were prepared by dilution from a stock solution (1000 mg l^{-1}) prepared using analytical grade ZnO (following a procedure recommended by the manufacturers of the AAS instrument used in this work); other chemicals used were of high-grade quality.

Removal of zinc by leaves was measured by following the decrease in concentration of zinc ions from solution in presence of a known amount of dry leaves. All experiments were carried out in air-conditioned room temperature ($\sim 20°C$). Clear solutions were obtained from the leaf suspensions, for the atomic absorption spectrometer (AAS) measurements, using a small glass tube with a sintered glass disc. Each experiment was done triplicate and the average of these readings was presented in the results. Readings with relative standard deviation above 10% were rejected. A quantity control sample, in the same concentration range of measured samples, was measured with each group of readings to check the accuracy of measurements. The method used for chemical analysis was the AAS (Pye Unicam, SP192). The pH of the solutions was monitored and adjusted to the desired value using 1 M NaOH or 1 M HCl, as required.

RESULTS AND DISCUSSION

Ability of Different Plant Leaves to Remove Zinc

Removal of zinc from 20 mg l^{-1} zinc aqueous solutions was studied using 15 types of plant leaves without pH adjustment. The results are shown in Figure 2(a) and (b). These results indicate that the efficiency of zinc removal by plant leaves varies considerably from one type of leaf to another. The best types of leaf for removing zinc at natural pH conditions are sisso, loquat, guava and walnut leaves. The least efficient types of leaf for the removal of zinc were olive and carob.

Effect of Acidity on the Removal of Zinc by Plant Leaves

Acidity has a large effect on the adsorption of metal ions on the various sorbents. This is largely because of the ability of hydrogen ions (with high density of charge and small ionic size) to compete with other metal ions on the adsorption sites. The effect of pH on the removal of zinc by the various types of leaves has been studied using pH values 2, 4 and 6. For this purpose pine leaves were selected and the results obtained on this sorbent are displayed in Figure 3. The efficiency of the removal process, after 150 h contact time, is shown in Table 2 at different pH values for the 15 types of plant leaves. The values of pH studied were limited to values below those causing precipitation of zinc hydroxide under the specified conditions used in this work (i.e. 7.3 based on calculations using K_{sp} for $Zn(OH)_2$ as 1.2×10^{-17}; Skoog and West, 1976). The results of Table 2 suggest that

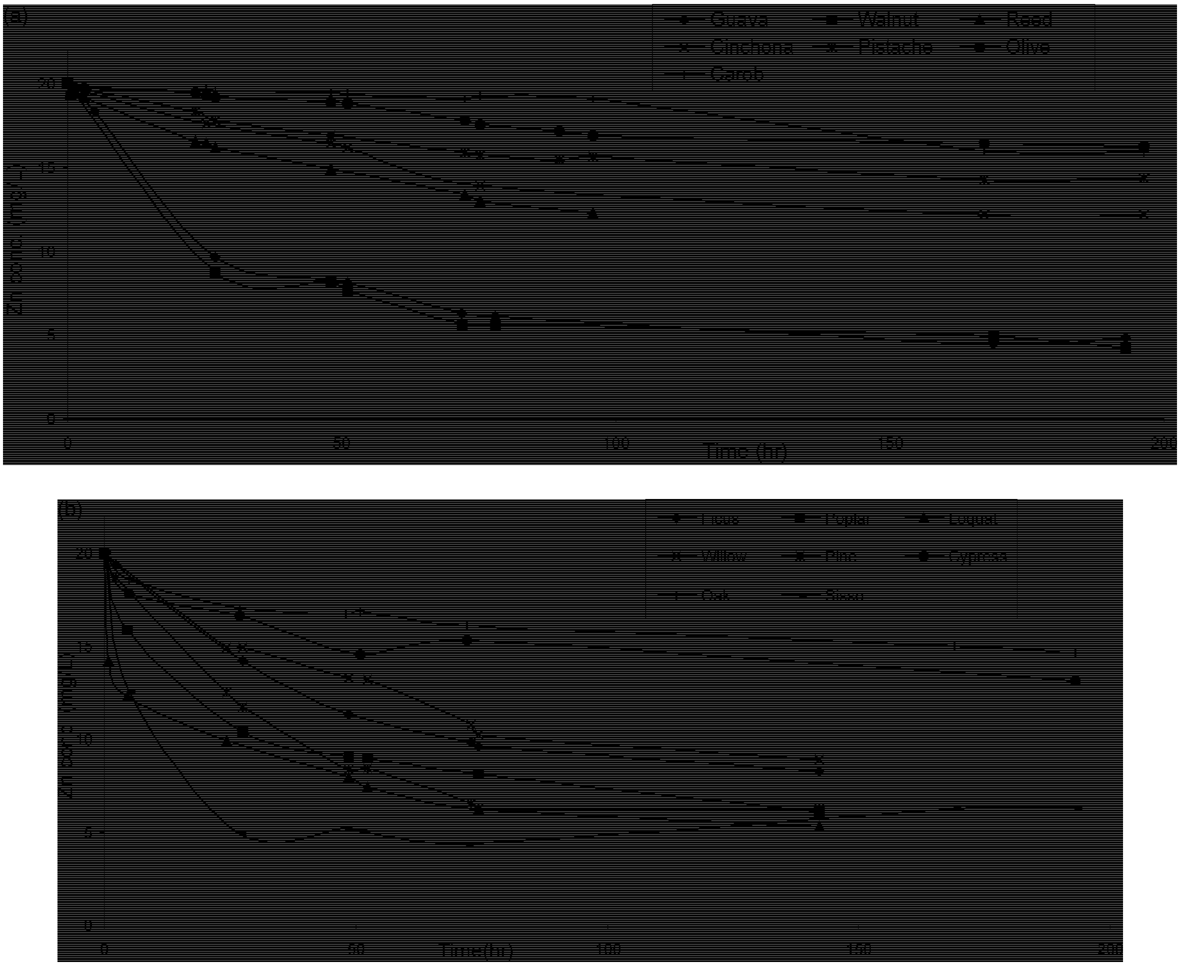

Figure 2. Removal of zinc from aqueous solutions using various types of plant leaves. Zinc concentration $= 20\,mg\,l^{-1}$, concentration of leaves $= 20\,g\,l^{-1}$, natural (unadjusted) pH.

walnut and poplar leaves are the most efficient plant leaves for the removal of zinc (~82%) from aqueous solutions at pH 6. At pH 2 the efficiency of the removal process was almost negligible. Previous studies showed different optimum values of pH for the removal of metal ions on plant leaves. For example, the best pH for removal of cadmium was 8.4 on cypress leaves (Sayrafi *et al.*, 1999), 5.9 on oak, pine and reed leaves (Sayrafi *et al.*, 1999) and ~5 on beech leaves (Salim *et al.*, 1992), and ~7 on *Cynodon dactylon* L. leaves (Salim and Abu-El-Halawa, 2002). The optimum pH for the removal of copper is 4.0 on poplar, walnut, reed, pistachio and guava leaves (Al-Subu *et al.*, 2001). It is 4.3 on sisso leaves (Al-Subu *et al.*, 2001), 6.0 on carob, cinchona, olive, ficus and oak leaves (Al-Subu *et al.*, 2001), 6.1 on cypress leaves (Al-Subu *et al.*, 2001), 6.2 on pine and loquat leaves (Al-Subu *et al.*, 2001) and ~5.4 on *Cynodon dactylon* L. leaves (Salim and Abu-El-Halawa, 2002). The optimum pH for the removal of nickel on pine leaves is 6.7 (Salim, 1988) and the optimum pH for the removal of lead on cypress leaves is 6.7 (Salim *et al.*, 1994).

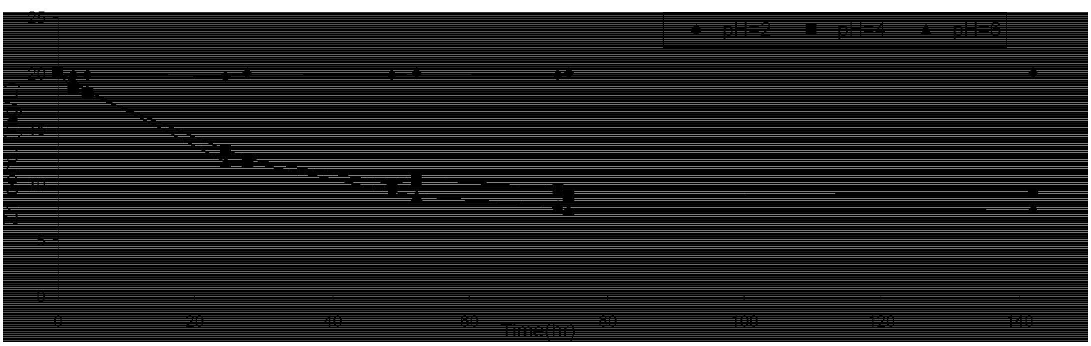

Figure 3. Effect of pH on the removal of zinc from $20\,mg\,l^{-1}$ aqueous zinc solutions using $20\,g\,l^{-1}$ pine leaves.

Table 2. Percentage of zinc ion removal from $20\,\mathrm{mg\,l^{-1}}$ zinc aqueous solutions at various pH values, after 150 h contact time.

	Percentage of Zn removal at different pH values		
Plant species	2	4	6
Olive	0.7	21.0	30.3
Reed	2.3	28.1	38.2
Carob	3.7	5.7	29.4
Willow	16.4	55.9	58.4
Loquat	25.0	67.1	74.8
Walnut	2.3	73.4	82.1
Sisso	7.2	49.5	71.5
Pine	0.3	54.0	61.1
Cinchona	2.0	37.8	51.9
Pistache	0.6	29.1	61.2
Oak	5.0	27.4	45.0
Ficus	28.1	57.6	63.4
Poplar	4.6	65.0	81.6
Cypress	6.9	27.5	42.2
Guava	3.5	52.3	78.3

Effect of Concentration of Zinc on Its Removal by Plant Leaves

Removal of zinc from various concentrations of zinc aqueous solutions, ranging from 2.0 to $20.0\,\mathrm{mg\,l^{-1}}$, was followed using fixed walnut leaves concentrations ($20\,\mathrm{g\,l^{-1}}$). The results obtained are given in Table 3. The amount of zinc ion removal increased with increasing zinc concentration in solution. The results were found to follow the Freündlich adsorption isotherm:

$$\log C_s = \log K + \frac{1}{n} \log C_l$$

where C_s = concentration of zinc in solution; C_l = concentra-ncentration of zinc on leaves; K and n = Freundlich constant parameters.

The parameters of this isotherm after 20 h contact time were calculated to be 0.76 and 468 for n and K, respectively. The parameter n is related to the sorption intensity and K is related to the sorption capacity of the sorbent. The measured parameters for zinc adsorption are very close to those reported for the adsorption of copper on plant leaves (Al-Subu *et al.*, 2001; Salim and Abu-El-Halawa, 2002). The parameter K obtained for zinc in the present work is considerably higher than values reported for the adsorption of lead (Salim *et al.*, 1994; Salim and Abu-El-Halawa, 2002) and of cadmium (Salim and Abu-El-Halawa, 2002; Salim *et al.*, 1992).

Effect of Concentration of Leaves on the Removal of Zinc

The effect of concentration of leaves on the removal of zinc from $20\,\mathrm{g\,l^{-1}}$ aqueous zinc solutions was studied using different concentrations of plant leaves ranging between 5 to $25\,\mathrm{g\,l^{-1}}$ dry walnut leaves at pH 4.0. The results obtained are shown in Figure 4. The results showed that increasing the concentration of leaves increases largely the efficiency of removal of zinc by leaves. The percentage removal of zinc by leaves increased linearly with the increase of the concentration of leaves up to a limit, after which this increase declined to almost a constant value. The maximum efficient

Table 3. Loss of zinc on dry walnut leaves ($20\,\mathrm{g\,l^{-1}}$) at pH 4.0.

	Original concentration of zinc ($\mathrm{mg\,l^{-1}}$)																			
	2.00		4.00		6.00		8.00		10.00		12.00		14.00		16.00		18.00		20.00	
Time (h)	C_s ($\mathrm{mg\,g^{-1}}$)	C_l ($\mathrm{mg\,l^{-1}}$)	C_s ($\mathrm{mg\,g^{-1}}$)	C_l ($\mathrm{mg\,l^{-1}}$)	C_s ($\mathrm{mg\,g^{-1}}$)	C_l ($\mathrm{mg\,l^{-1}}$)	C_s ($\mathrm{mg\,g^{-1}}$)	C_l ($\mathrm{mg\,l^{-1}}$)	C_s ($\mathrm{mg\,g^{-1}}$)	C_l ($\mathrm{mg\,l^{-1}}$)	C_s ($\mathrm{mg\,g^{-1}}$)	C_l ($\mathrm{mg\,l^{-1}}$)	C_s ($\mathrm{mg\,g^{-1}}$)	C_l ($\mathrm{mg\,l^{-1}}$)	C_s ($\mathrm{mg\,g^{-1}}$)	C_l ($\mathrm{mg\,l^{-1}}$)	C_s ($\mathrm{mg\,g^{-1}}$)	C_l ($\mathrm{mg\,l^{-1}}$)	C_s ($\mathrm{mg\,g^{-1}}$)	C_l ($\mathrm{mg\,l^{-1}}$)
4	0.007	1.86	0.020	3.60	0.040	5.20	0.060	6.80	0.073	8.53	0.09	10.20	0.10	12.00	0.10	14.00	0.113	15.75	0.130	17.40
8	0.014	1.72	0.045	3.10	0.080	4.40	0.112	5.75	0.140	7.20	0.17	8.52	0.20	10.00	0.23	11.40	0.250	13.00	0.275	14.50
12	0.020	1.60	0.065	2.70	0.115	3.70	0.152	4.95	0.200	6.00	0.24	7.20	0.28	8.40	0.32	9.60	0.327	11.45	0.370	12.53
16	0.030	1.38	0.090	2.20	0.150	2.95	0.200	4.00	0.250	4.95	0.32	2.58	0.37	6.53	0.40	8.00	0.453	9.30	0.481	10.38
20	0.040	1.20	0.107	1.86	0.186	2.28	0.250	2.95	0.320	3.60	0.40	4.00	0.46	4.80	0.54	5.20	0.550	7.00	0.600	8.00

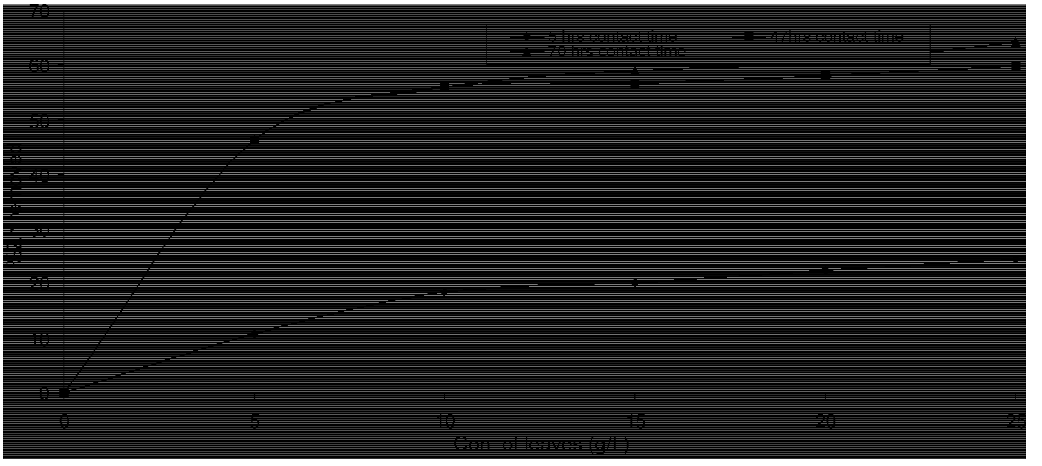

Figure 4. Effect of concentration of leaves on the percentage of zinc removal from $20\,mg\,l^{-1}$ aqueous zinc solutions, pH 4.0, on walnut leaves.

concentration of leaves for the removal of zinc is about $15\,g\,l^{-1}$.

Effect of Agitation

The effect of agitation on the removal of zinc was studied using $20\,mg\,l^{-1}$ of zinc aqueous solutions and $20\,g\,l^{-1}$ of walnut leaves at pH 4.0. The results (Figure 5) indicated that agitation increased the efficiency of the removal process. This conclusion is in agreement with previous findings on the removal of lead (Salim *et al.*, 1994) and of copper by plant leaves (Al-Subu *et al.*, 2001).

Effect of Competing Ions

The effect of the presence of competing ions on the removal of zinc was studied using $20\,mg\,l^{-1}$ zinc aqueous solutions and $20\,g\,l^{-1}$ walnut leaves. The ions studied were Ag, Ca, Cd, Cu, Mg, Na, Ni and Pb. The concentration of each of these competing ions was $100\,mg\,l^{-1}$ and the pH was adjusted to 4.0. The results (Figure 6) indicated that presence of Ca, Cd, Cu, Mg, Ni and Pb reduced the efficiency of plant leaves to remove zinc from solution. This effect was in the decreasing order: Pb > Cd > Mg > Ca > Cu > Ni. On the other hand, the presence of Na or Ag ions in solution increased slightly the efficiency of walnut plant leaves in removing zinc from aqueous solutions.

Foreign ions are expected to affect the removal process of zinc by competing with zinc on the adsorption sites of leaves. The most effective competing ion is the hydrogen ion because of its small size and high charge density. Hydrogen ions showed a large influence on the adsorption of zinc as discussed earlier in this work. The effect of the Pb, Cd, Mg, Ca, Cu and Ni ions on reducing the efficiency of the removal of zinc might be explained as due to the competition with zinc on the vacant adsorption sites. On the other hand, the effect of Na^{+} and Ag^{+} ions which resulted in increasing adsorption of zinc on plant leaves is more difficult to explain than above. A possible explanation might be that these ions are capable of occupying new adsorption sites and making them available for zinc by interaction between these ions and zinc ions. This positive effect of the presence of some ions was shown in other previous studies. Examples are the effect of lead on increasing the removal of cadmium (Salim *et al.*, 1992), the effect of Na^{+} on increasing the removal of lead (Salim *et al.*, 1994) and the effect of Na^{+} Zn^{+} and Mg^{2+} on increasing the removal of copper (Al-Subu *et al.*, 2001).

Effect of Presence of EDTA

The effect of presence of the complexing agent EDTA on the removal of zinc from $20\,mg\,l^{-1}$ aqueous zinc solutions on $20\,g\,l^{-1}$ walnut leaves was studied using various concentrations of EDTA ($10–100\,mg\,l^{-1}$) at pH 4.0. The results

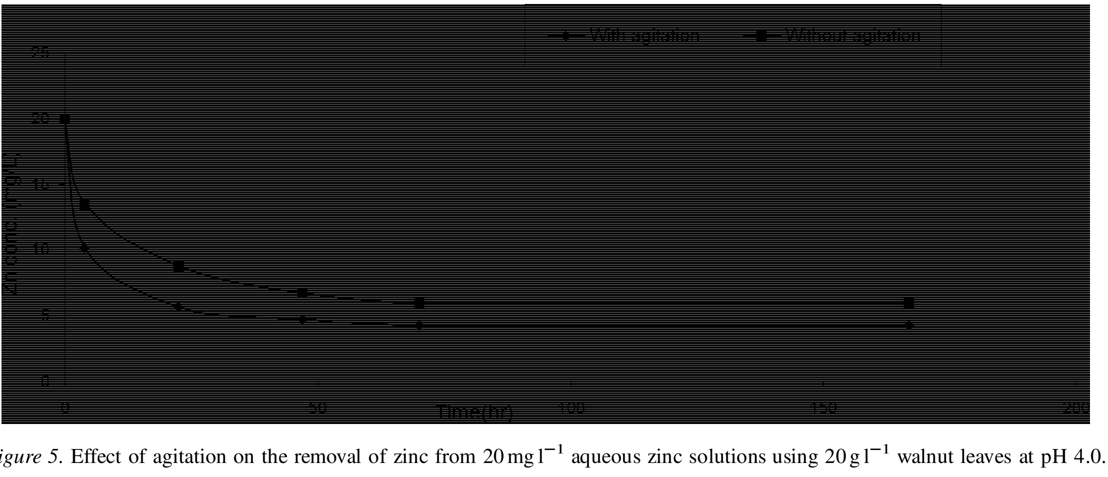

Figure 5. Effect of agitation on the removal of zinc from $20\,mg\,l^{-1}$ aqueous zinc solutions using $20\,g\,l^{-1}$ walnut leaves at pH 4.0.

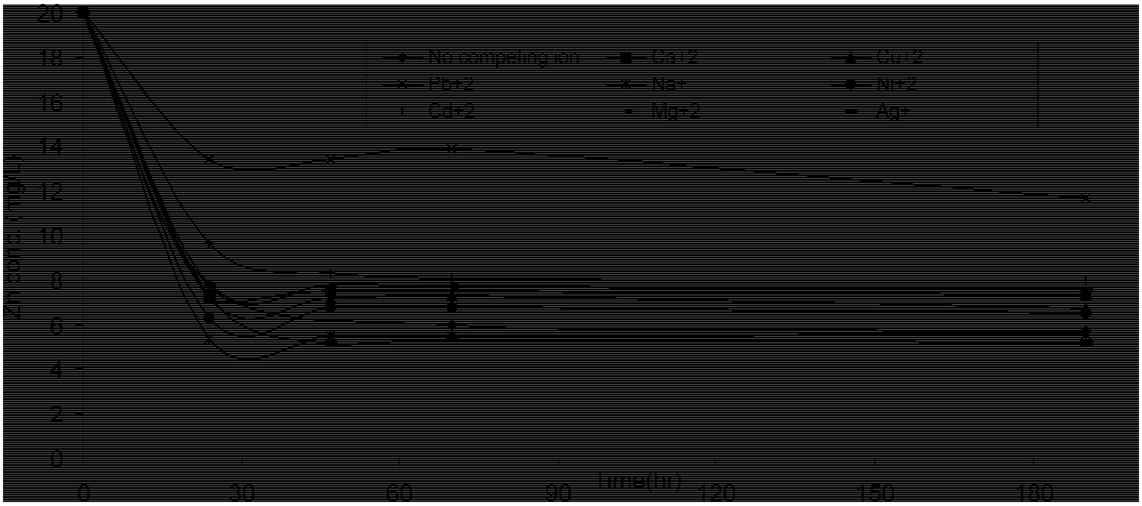

Figure 6. Effect of presence of competing ions ($100\,mg\,l^{-1}$) on the removal of zinc solutions on $20\,g\,l^{-1}$ walnut leaves at pH 4.0.

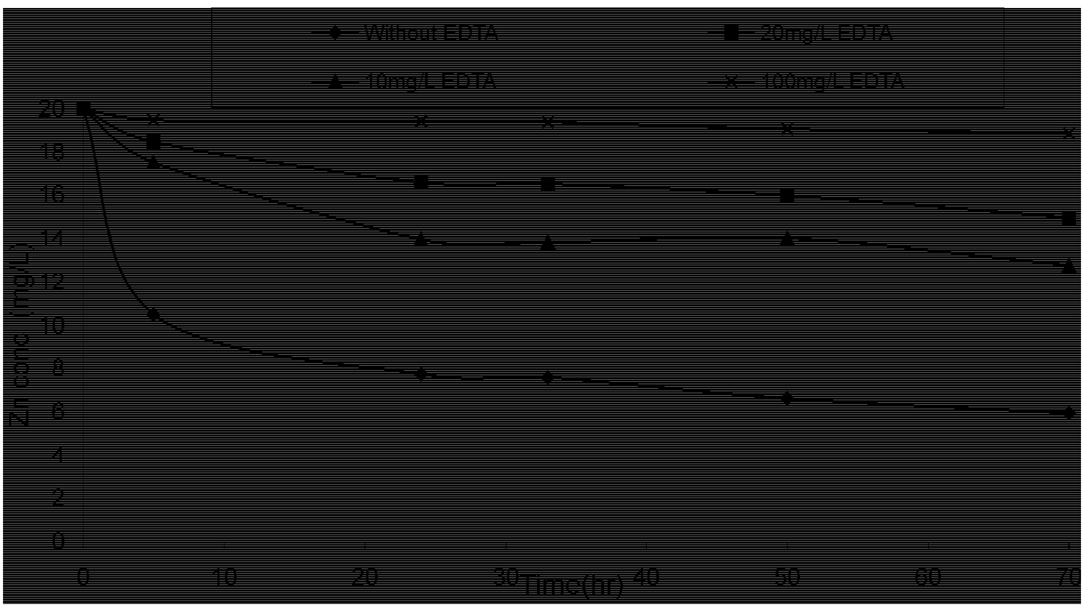

Figure 7. Effect of presence of EDTA on the removal of zinc from $20\,mg\,l^{-1}$ aqueous using $20\,g\,l^{-1}$ walnut leaves at pH 4.0.

obtained (Figure 7) indicated that increasing EDTA concentration in the solution greatly reduced the efficiency of zinc removal. This suggests that the efficiency of removal of zinc from aqueous solutions depends largely on the state of ions, free ions being much easier to adsorb onto plant leaves than complexed ions. This might be attributed to the smaller size of free ions than complexed ions. The present results agree with previous studies on the effect of EDTA on the removal of lead (Salim *et al.*, 1994) and of copper by plant leaves (Al-Subu *et al.*, 2001).

CONCLUSIONS

Plant leaves were found capable of removing zinc ions from aqueous solutions. The removal efficiency of zinc was found dependent on the plant species used. The best types of leaves for removing zinc from natural (unadjusted pH) solutions were sisso, loquat, guava and walnut leaves.

Acidity of solution affects largely the efficiency of removal of zinc from aqueous solutions. The maximum efficiency for removal ($\sim$82%) was at pH $\sim$6 using walnut or poplar leaves.

The amount of zinc removal from solution increased with increasing zinc concentration in solution. The results showed the applicability of the Freündlich adsorption isotherm. The removal efficiency of zinc was increased with increasing concentration of plant leaves up to a limit of $\sim 15\,g\,l^{-1}$.

The presence of competing ions in solution reduced the efficiency of the removal process except sodium and silver ions, which increased slightly the efficiency of the removal process. The presence of EDTA reduced the efficiency of the removal process. Agitation of solutions increased the efficiency of the removal of zinc by plant leaves.

REFERENCES

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

Al-Asheh, S. and Duvnjak, Z., 1997, Adsorption of metal ions by moss, *Adv Environ Res*, 2: 194–212.

Al-Subu, M., Salim, R., Abu-Shqair, I., Braik, H. and Swaileh, K., 2001, Removal of dissolved copper from polluted water using plant leaves: I. Effect of acidity and plant species, II. Effects of copper concentration, plant leaves, competing ions and other factors, *Rev Int Contam Ambient*, 17: 91–96, 123–127.

Brooks, R.R., 1978, Pollution through trace elements, in *Environmental Chemistry*, Bockris, J.O.M. (ed.) (Plenum Press, New York, USA), pp 429–477.

Dojlido, R. and Best, G., 1993, *Chemistry of Water and Water Pollution* (Ellis Horwood, New York, USA), pp 79–83.

Gin, K.Y., Tang, Y. and Aziz, M.A., 2002, Derivation and application of a new model for heavy metal bio-sorption by algae, *Water Res*, 36: 1313–1323.

Hammond, P.B. and Beliles, R.B., 1980, in *Toxicity: the Basic Science of Poisons*, Casarett, C. and Doull, B. (eds) (MacMillan, New York, USA).

Kobayashi, R. and Nishi, S., 1974, Removing heavy metals from wastewater by treating with modified wool or silk, *Jap Kak*, 74: 16675.

Lee, T.A. and Hardy, J.K., 1987, Copper uptake by water hyacinth, *J Environ Sci Health*, A22: 141–160.

Lee, S.H., Chong, H.J., Hongsuk, C., Moo, Y.L. and Won, Y.J., 1998, Removal of heavy metals from aqueous solutions by apple residues, *Proc Biochem*, 33: 205–211.

Moore, J. and Ramamoorthy, S., 1984, *Heavy Metals in Natural Waters* (Springer, Berlin, Germany).

Ornes, W.H., 1994, Bioaccumulation of cadmium in duckweed, *J Environ Sci Health*, A29, 1035–1044.

Salim, R., 1988, Removal of nickel (II) from polluted water using decaying leaves, *J Environ Sci Health*, A23: 183–197 and 321–334.

Salim, R. and Abu-El-Halawa, R., 2002, Efficiency of dry plant leaves (mulch) for removal of lead, cadmium and copper from aqueous solutions, *Trans IChemE*, *Part B*, *Proc Safe Env Prot*, 80B: 270–276.

Salim, R. and Hilal, H.S., 1986, Conditions for minimum adsorption and zinc container surfaces, *J Environ Sci Health*, A21: 681–690.

Salim, R. and Robinson, J.W., 1985, Removal of dissolved aluminum released by acid rain using decaying leaves, *J Environ Sci Health*, A20: 701–734.

Salim, R., Al-Subu, M. and Sahrhage, E., 1992, Uptake of cadmium from water by beech leaves. *J Environ Sci Health*, A27, 603–627.

Salim, R., Al-Subu, M. and Qashoa, S., 1994, Removal of lead from polluted water using decaying plant leaves, *J Environ Sci Health*, A29: 2087–2114.

Sayrafi, O., Salim, R. and Sayrafi, S., 1999, Removal of cadmium from polluted water using decaying leaves, *J Environ Sci Health*, A34: 835–851.

Singh, D.K., Tiwari, D.P. and Saksena, D.N., 1993, Removal of lead from aqueous solutions by chemically treated tealeaves, *Ind J Environ Health*, 35: 169–177.

Skoog, D.A. and West, D.M., 1976, *Fundamentals of Analytical Chemistry*, 3rd edn (Holt Rinehart and Winston, New York, USA), Appendix 3.

Sobhan, R. and Sternberg, S., 1999, Cadmium removal using Cldophora, *J Environ Sci Health*, A34: 53–72.

Wilhelm, M., Ohnesorge, F.K., Lombeck, I. and Hafner, D., 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.

ADDRESS

Correspondence concerning this paper should be addressed to Dr R. Salim, Chemistry Department, An-Najah University, Nablus, PO Box7, West Bank, Palestine.
E-mail: radisalim@yahoo.com

The manuscript was communicated via our Regional Editor for Asia Dr Amer El-Hamouz. It was received on 24 April 2002 and accepted for publication after revision 30 May 2003.