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VARIATION OF METALS IN BED SEDIMENTS OF QARAAOUN RESERVOIR, LEBANON

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Abstract. The Qaraaoun Reservoir (impoundment of the River Litani) is the only artificial surface water body in the country, Lebanon. Earlier study on the water quality of the Qaraaoun Reservoir identified three water quality zoning with a central distinct zone suitable for multipurpose water usage. The objective of this study was to extend the earlier work by considering the total metal content of reservoir bed sediments and hence to evaluate factors that control metal deposition or capture. Water samples were collected from 15 sampling sites and sediment samples were simultaneously collected from 9 sites. Water parameters analyzed were pH, Eh, DO and temperature. Sediment samples were dried and sieved and sediment $<75\ \mu m$ was retained for analysis. Sediments were subjected to a stepwise heating process with aqua regia to extract the metals, and their content in sediments determined by ICP-MS. The sediment data revealed higher metal contents where the river entered the reservoir which matched higher concentrations of water parameters at the influx site. Regression analysis of total metals in sediments with distance from the river Litani influx point to the dam revealed a log trend for Fe, Cr and Ni, whereas, the concentrations of Cu, Zn, Cd, Pb were better described by a polynomial regression. Three sediment zones were identified: entrance, oxidation (central) and reducing (near dam) zones. Sediment contents of Zn, Cu and Pb correlated with organic content, whereas sediment Cr and Ni were associated with iron. It was concluded that sediments act as a sink for metals and the deposition of metals is primarily related to sediment organic content and the level of dissolved oxygen in water.

Keywords: metals, sediment, Qaraaoun reservoir, Lebanon

1. Introduction

Lebanon posses relatively abundant water supply compared to its neighboring countries. Reliance on its major water resource, the ground water is impaired by quality deterioration and excessive utilization (Jurdi, 1992, 1998; Khair *et al*., 1994; Sene *et al*., 1999; El-Fadel *et al*., 2000). More attention must be paid now to managing and using surface water (Jurdi *et al*., 2002). Technical reports have recommended developing a network of 16–20 dams distributed through out the country (Amery, 2002). At present, the Qaraaoun Reservoir (impoundment of the River Litani) is the only artificial surface water body in the country. Previous study (Jurdi *et al*.,

2002) on the water quality of the Qaraaoun Reservoir was evaluated and a distinct zone within the reservoir was identified for multipurpose water usage. Such type of water quality assessment is inadequate for a long term management. As such, the assessment of sediment quality is of utmost importance. Sediments are the sink of metals in fresh and marine environment (Louma, 1989; Arjonilla *et al.*, 1994; Weimin *et al.*, 1994, Hudson-Edwards *et al.*, 1997; Large and McGoldrick, 1998, Korfali and Davies, 2000; An and Kampbell, 2003). Sediments are important components of lake ecosystems in which toxic compounds accumulate through complex physical and chemical adsorption mechanisms depending on the nature of sediment matrix and the properties of the adsorbed compounds (Ankley *et al.*, 1992; Maher and Aislabie, 1992; Leivouri, 1998). The adsorption process involves a dynamic exchange between the adsorbed materials and water and is influenced by several physical and chemical parameters like: pH, oxidative-reductive potentials, dissolved oxygen, organic and inorganic carbon content (De Bartolomeo *et al.*, 2004). Thus, the deposition of metals in sediments occurs through an interaction between sediment and water (Piron *et al.*, 1990), whereby variations of metal contents of sediments and water depends on variation of water chemistry for example temperature, pH and solute concentration (Korfali and Davies, 2003). Besides, bed sediment metal contents reflect the influence of catchment lithology and anthropogenic contamination. Transport, deposition, re-suspension and solubilisation of these metals in the fluvial system is dominated by hydrological processes and by the chemistry of the water column: there are many gaps in our understanding of this complex system (Foster and Charlesworth, 1996). The objective of this study is to support the previous study (water quality) by considering (determine) the total metal content of reservoir bed sediments. Hence, to evaluate factors that control metal deposition or capture and comprehensively evaluate the Qaraaoun water reservoir.

2. Study Area

The Qaraaoun Reservoir is an impoundment of Litani River located in Bekaa valley that lies between the two Lebanese mountains Ranges (Mount Lebanon and Anti-Mount Lebanon) that outcome a semi-arid climate due to the shut of the Bekaa Valley from the tempering effect of the sea climate (Sene *et al.*, 1999; Jurdi *et al.*, 2002). The Litani River is the longest (170 km) and largest river in Lebanon. The river's source is the Al Oliek spring in the Bekaa Plain (west of Baalbeck) and flows southward parallel to Mount Lebanon and discharges into the Mediterranean Sea, 7 km north of Tyre. Geomorphologically, the Litani basin is divided into three sub-basins, the largest is the upper one stretching from the Bekaa Plain to Qaraaoun Dam (built between 1958–1965). The average annual discharge of Litani River in this sector is approximately 13.02 m^3/s (UNDP, 1970; Khair *et al.*, 1992; Jaber, 1993). The Dam is 110 m long and 61 m high forming the Qaraaoun Reservoir that can store up to 220 million m^3 of water, 160 million m^3 of which are used annually for irrigation, industry and hydropower, and 60 million m^3 remain in storage throughout the dry seasons (Khair, 1993; Owaydah, 1993; Jurdi *et al*., 2002).

Geologically, the rocks outcropping in the Qaraaoun Reservoir basin belong to the Jurassic, Cretaceous, Tertiary and Quaternary system (Figure 1). Most of the rocks of the Jurassic system (J_6), Cenomanian (C_4) and Eocene (e_{2b}) are limestone and dolomitic limestone. In a few localities, Conomanian rocks (C_3 and C_6) outcrop consisting mainly of chalky marl. The Quaternary deposits (q), are of limited extent and comprise mainly alluvial deposits consisting of clay, silt, sand and gravel.

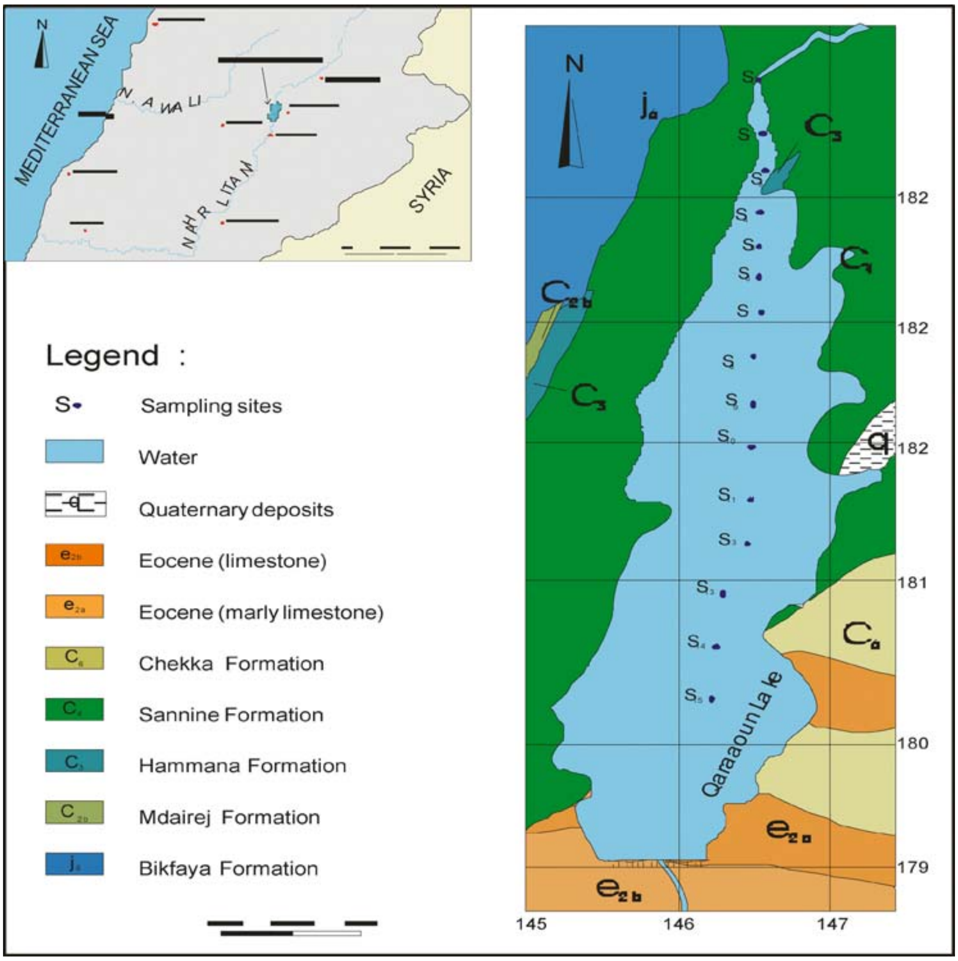

Figure 1. Geological map of Qaraaoun Reservoir and location of sampling sites.

3. Materials and Methods

3.1. Sampling sites

The river Litani valley north of Qaraaoun Reservoir was inventoried prior to water and sediment sampling to identify possible types of contaminates entering the reservoir. Major contaminant sources comprise: sewage network outlets, municipal solid waste dumping, industrial zone (dyeing and tanning, electroplating, manufacturing batteries, sponge and paper), food processing plants, farms (poultry, cows and sheep) and agricultural run offs (Jurdi *et al.*, 2002).

Similar to the earlier work done in the year 1995, sampling was carried out in the dry season (August, September, November, October, 1998). This would reflect on minimal dilution of water born pollutants. Water samples were collected from 15 sampling sites and sediments were simultaneously from 9 sites within the reservoir. The last site (near dam) is restricted by the accessibility of reach (Figure 1).

3.2. Field methods

Water samples for analysis were collected at each sampling site at mid depth from the surface using two 1L polyethylene bottles. The polyethylene bottles were presoaked overnight in 10% (v/v) nitric acid and then rinsed with distilled water. Before filling with the sample they were also rinsed twice with water resource. The method of sampling and collection are in accordance with Standard Methods by American Public Health Association, American Water Works Association, Water Pollution Control Federation (APHA, AWWA, WPCF, 1992). Temperature, electrical conductivity (EC_w), total dissolved solids (TDS), pH and Eh were determined immediately after sampling using (Hach Model 44600 Conductivity/TDS Meter, resolution Conductivity 0.1 μS/cm, TDS 0.1 mg/L).

Sediments (grab) were collected using gravity corer method into polyethylene container according to standard methods (Chapman, 1992). The pH and Eh were determined immediately after sampling.

3.3. Laboratory anlysis

Sediment samples were dried at room temperature and sieved, the sediment size $<75\ \mu$m size was retained. Total metal in sediment was extracted with hot aqua regia (Kersten and Förstner, 1989) and amounts of extracted metals then determined by ICP-MS techniques. The inorganic content in sediments were determined by back titration method (Nelson, 1982), whereas organic content by LOI method (Davies, 1974).

TABLE I
Water and sediment parameter related to water sediment interaction

Site	pH (water)	pH (sediment)	DO (water) mg/L	Eh (sediment) mv	OC (sediment) %	INC (sediment) %
1	7.60	7.20	5	58	5.17	5.40
2	7.50	7.50	6	53	2.80	6.2
3	7.90	7.60	7	75	1.40	10.01
4	8.10	7.10	7	128	1.21	10.20
5	8.10	7.20	8	133	1.05	9.50
6	7.80	7.60	8	113	0.86	7.10
7	8.10	8.10	6	67	1.30	6.80
8	8.00	8.00	5	20	1.87	5.30
9	7.90	7.90	4	−56	2.15	2.00

OC = Organic carbon, INC = Inorganic carbon (% C-CO3)

4. Results and Discussion

4.1. pH, Eh WATER AND SEDIMENT

Parameters that relate to metal sediment interaction and sediment quality are presented in Table I. The pH of water ranged from (7.5–8.1) and those of sediment (7.1–8.1) and this reflects that the Qaraaoun water and sediments are alkaline which is typical of water bodies underlain by carbonate rocks (Korfali and Davies, 2000). The saturation pH is 8.3 for fluvial system open to atmospheric CO_2 having a partial pressure of $10^{-3.5}$ bar and saturated with calcium ion (Stumm and Morgan, 1996). Fluvial system affected by mining activities, as well as acidic effluents outcome acidic water conditions. As such, the Qaraaoun reservoir is not receiving acidic effluents and this is confirmed by the absence of reported mining activities in Lebanon. In addition, it could be experiential that the Qaraaoun reservoir whose basin is underlain by carbonate rocks exhibit high pH. Thus, it has the capacity to lower the de-sorption of metals from sediments and to possess high buffering capacity against acidic inputs (Jarvie *et al.*, 1997). The Eh values in sediments ranged from oxidizing conditions to reducing conditions near the dam. These values are in parallel with the amount of dissolved oxygen (DO) content in water. Constructions of dams in water bodies render poor water quality due to the induced anoxic conditions (Rast and Thorton, 1999). The highest values of dissolved oxygen were noted in the central part of the reservoir. This central part is the widest and shallowest segment (Figure 1); oxygen recovery is more enhanced in such localities (Chapman, 1992). The effect of Eh values and dissolved oxygen affected the overall amounts of organic carbon content in sediments (Table I). The prevailed oxidized condition lowered the organic carbon content and the prevailed reduced condition elevated the organic carbon content.

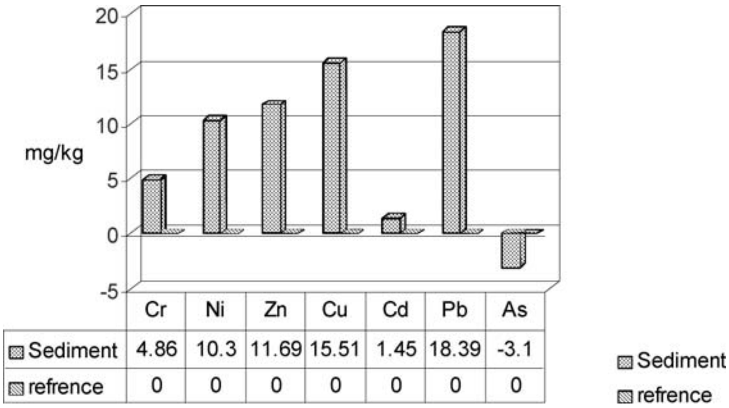

Figure 2. Difference of levels of mean metal content to reference (limestone).

4.2. METALS IN SEDIMENTS

The levels of metals in Qaraaoun Reservoir sediments were compared to the metal content in the limestone rocks after Li (2000). Figure 2 depicts the difference of the mean sediment metal content and that of the reference (limestone metal content).

It is observed that the reservoir sediments have metals content higher (except As) than their back ground reference. This indicates that the reservoir is subjected to enrichment by other sources than its natural setting environment. However, the variation of the metals content within the reservoir (from entrance to dam) would asses precisely the sediment quality which in return reflects on the water quality. This will ultimately lead to a re-justification of the previous proposed zoning intended for multi purpose water utilization.

All measured metals have their highest content in sediments at the entry of the reservoir (influx point). This is in line with the earlier work reporting on water quality (Jurdi *et al*., 2002). The influx site receives contaminants form Litani valley north of Qaraaoun Reservoir that was inventoried prior to water and sediment sampling, and accordingly possible types of contaminates entering the reservoir were identified. Major contaminants sources were characterised by: sewage network outlet, municipal solid wastes, food processing plants, industrial zone, farms and agricultural (Jurdi *et al*., 2002).

Regression analysis of total metals content in sediments with distance from Litani influx point to dam revealed a log trend for Fe, Cr and Ni (Figure 3); whereas Cu, Pb, Zn, and Cd were better described by a polynomial regression (Figure 4). Regression analysis of these metals predicted the best-fit mathematical equation to be polynomial form and not linear relation. Three zones along the water reservoir were depicted to regulate metal deposition: entrance, oxidation and reduction zones and based upon measured pH, Eh of sediment and DO content of water (Figure 5).

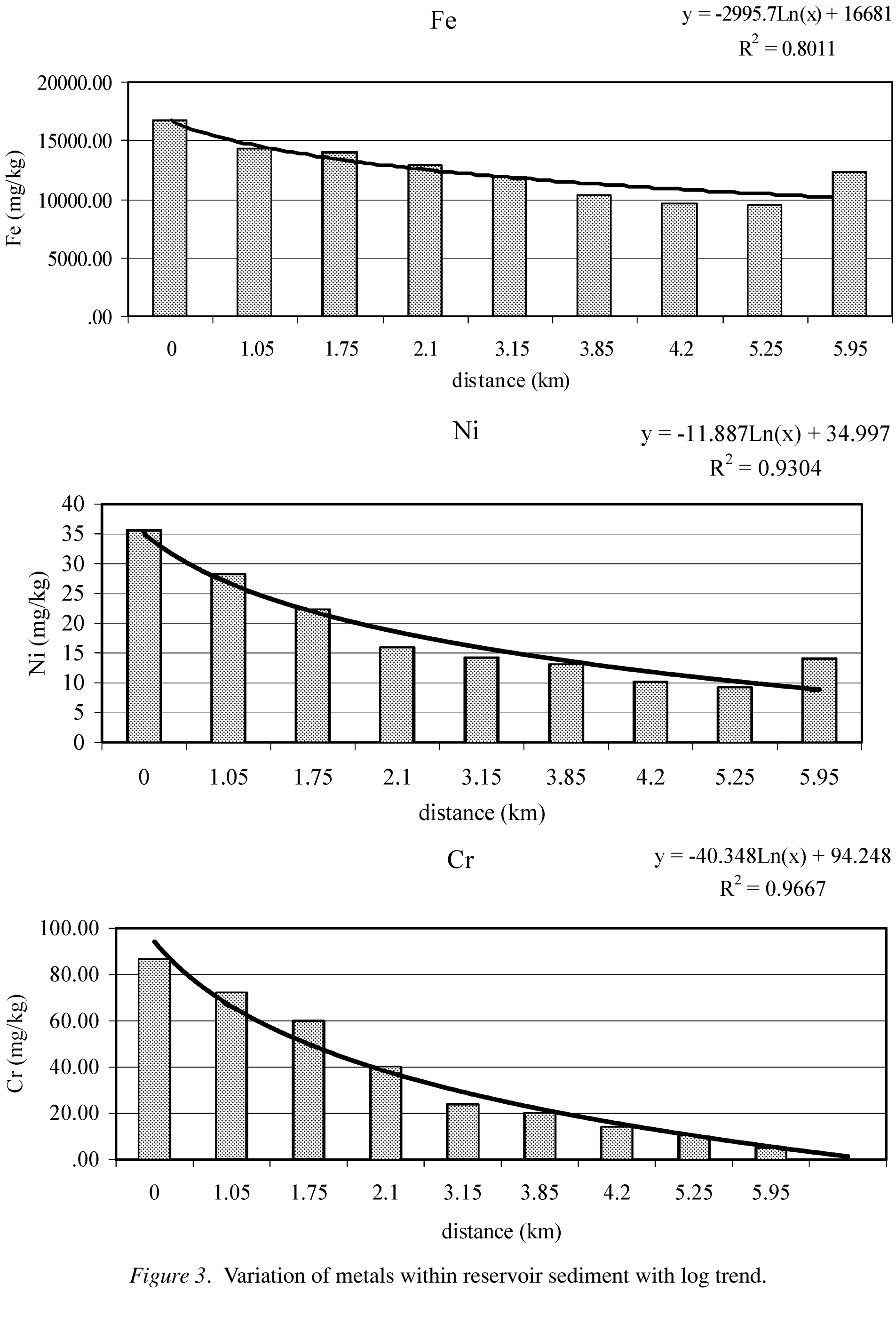

Figure 3. Variation of metals within reservoir sediment with log trend.

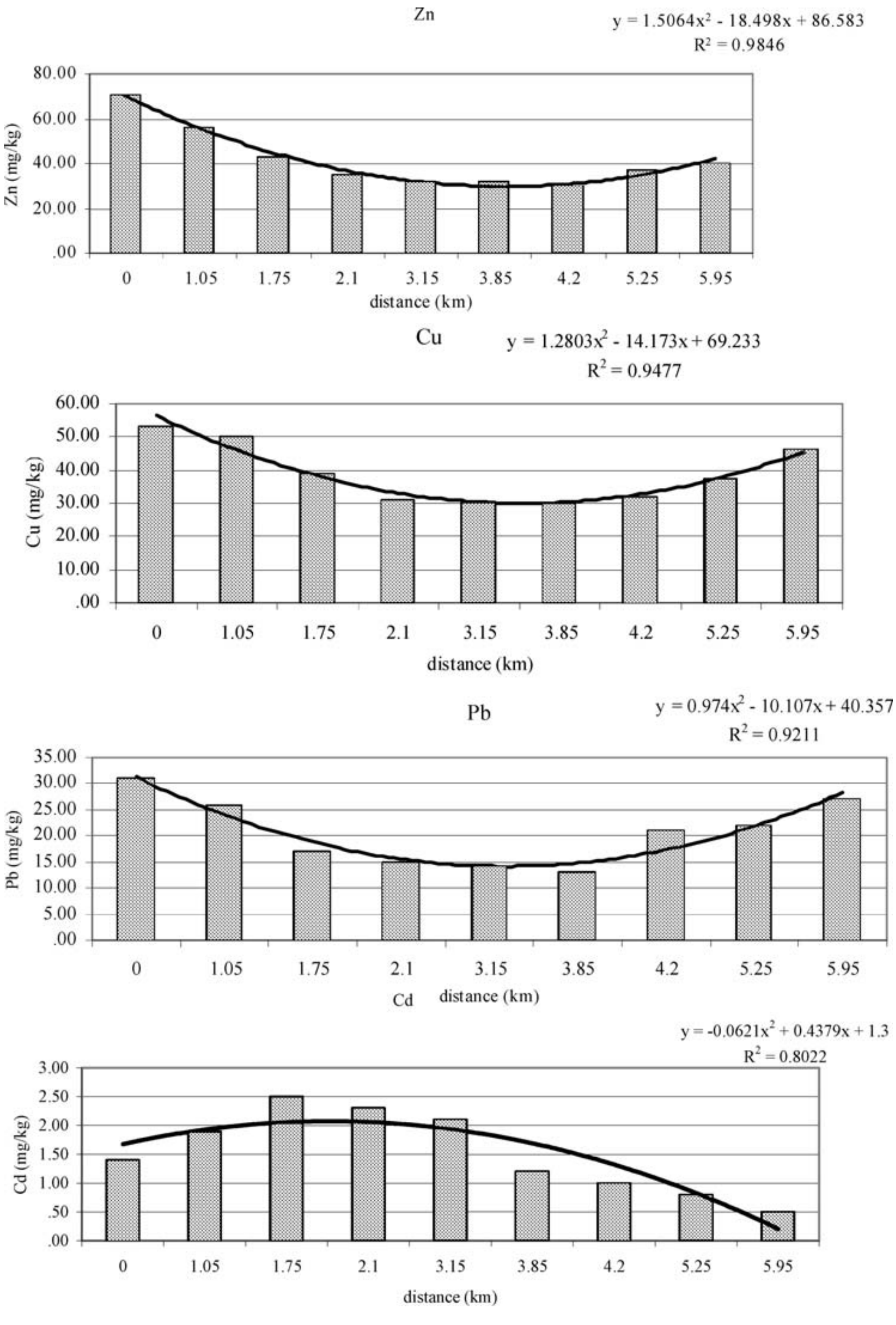

Figure 4. Variation of metals within reservoir sediments with polynomial trend.

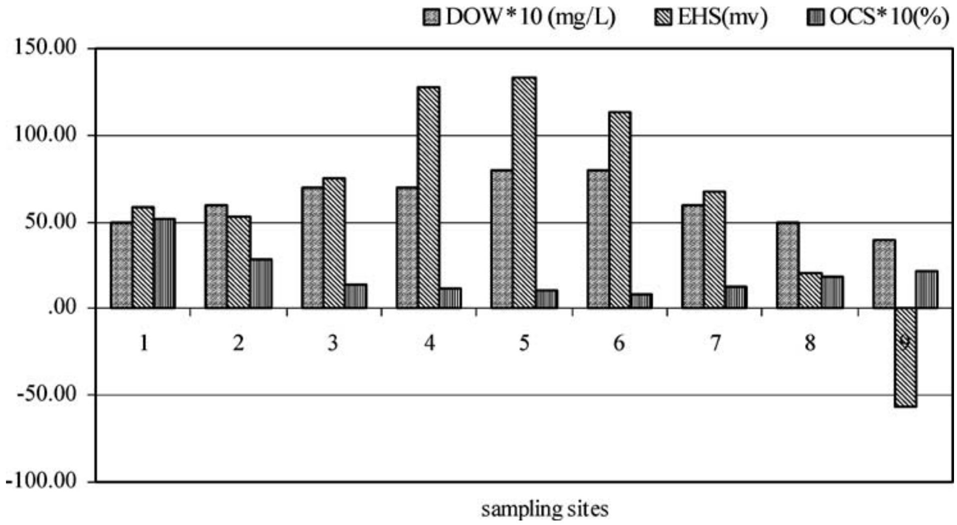

Figure 5. Zoning within Qaraaoun Reservoir affecting metal sediment deposition.

4.3. Iron, chromium and nickel

Iron, Cr and Ni showed the same trend line variation within the reservoir (Figure 3). The high content of Cr and Ni in sediments at the influx point is most probably due to the industrial wastes discharges of the upper Litani Valley industries (tanneries, electroplating, and battery factories). These industries are major sources of chromium and Ni (Stephenson *et al*., 1998; Stępniewsks and Bucior, 2001).

The sediment content of Cr and Ni was related to Fe sediment content. No relation occurred with content of sediment organic carbon. Statistical significant Pearson correlation was exhibited between sediment Fe content and those of Cr and Ni; no such correlation occurred with respect to sediment organic content (Table II). Hursthouse *et al*. (2001, 2003) have reported the strong correlation between total Fe content and Cr content in sediments. Iron oxides reveal in sediments and soil the highest affinity for adsorption of Cr ensuing a stronger relationship than those of sediment clay and organic content (Kabata-Pendias, 2000). This high association with Fe sediment could explain the lower values of Cr and Ni near the dam. A

TABLE II
Pearson correlation coefficient

	Ni	Cr
Fe (sediment)	0.947*	0.894*
% OC (sediment)	0.548	0.441

$^*p < 0.01$.

reduction condition reduces the insoluble ferric into the soluble ferrous in sediments and as a consequence expelling the Cr and Ni.

4.4. Zinc, copper and lead

Similarly, the high content of Zn, Cu, Pb and in sediments at the influx point is most probably due to industrial wastes discharges of the upper Litani Valley industries previously noted. The sediment content of these metals was related to the organic sediment content. Statistical significant Pearson correlation was exhibited between sediment Zn, Cu and Pb with sediment organic content and significant negative correlation with levels of dissolved oxygen in water; no such correlation occurred with respect to sediment Fe content except for Zn (Table III). Organic matter is considered as an important sink of Zn, Cu, and Pb in soils (Kabata-Penduis, 2000; Mellor, 2001; Wong *et al.*, 2002; Díaz-Barrientos *et al.*, 2003). The high association with organic matter could clearly explain the polynomial regression of these metals within the reservoir (Figure 4). The presence of higher content of dissolved oxygen in water in the central zone (Figure 5) induced an oxidation of the organic sediment with a consequence of expelling out the associated metals. However, the higher content, near dam or exit, of metals in sediment are most probably due to the higher organic content in sediments resulting from the prevailing reducing conditions (Table III, Figure 5). Lower levels of metal content near the dam would be expected for the metals that are associated with the Fe sediment phase. Nevertheless, Zn showed exception in its correlation with Fe sediment phase. This observation could be explained that both phases of sediments the organic and Fe regulate Zn geochemistry.

4.5. Cadmium

The variation of Cd sediment content along the reservoir revealed a different pattern. The variation was polynomial (convex shape rather than concave). High Cd sediment content was noted in the central zone of reservoir (Figures 4 and 5). Cadmium did not show association with organic content, nor with Fe phase (Table IV). Statistical significant Perason correlation occurred between Cd content and inorganic

TABLE III
Pearson correlation coefficient

	Cu	Pb	Zn
%OC (sediment)	0.879**	0.866**	0.950**
DO (water)	−0.695*	−0.875*	−0.441
Fe (sediment)	0.458	0.489	0.858*

*$p < 0.05$; **$p < 0.01$.

TABLE IV
Pearson correlation coefficient

	%OC (sediment)	Fe (sediment)	%INC (sediment)	pH (sediment)
Cd	−0.153	0.458	0.876**	−7.07*

$^{*}p < 0.05$; $^{**}p < 0.01$.

carbon content in sediments. It is concluded that what regulates the geochemistry of Cd in sediment is the carbonate sediment phase. This association of Cd with carbonate sediment phase was discussed in details in previous works (Korfali and Davies, 2000, 2003).

5. Conclusion

Previous study on water quality assessment of the Qaraaoun Reservoir identified a distinct oxidation zone (middle) suitable for multipurpose utilization. This study extended the previous work by considering total metal content of reservoir bed sediment. In parallel three zones within the reservoir were identified to regulate the metal deposition in sediment. Influx zone (the entrance of Litani River to reservoir) had the highest content of metals. An oxidation zone (middle) reduced the organic sediment content and decreased the content of metals (Cu, Pb and Zn) that were noted to be regulated by the organic content. A reduction zone (near the dam), reduced the iron content in sediment with a consequence decrease of metals (Cr and Ni) regulated by Fe sediment phase. The content of Cd in sediment was related to carbonate sediment phase. The overall process of metal sediment deposition in the reservoir would be improved by subjecting the sediments to sequential techniques of metals (fractionation-in hand), XRD and microprobe analysis of sediments.

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