

**Heavy metals pollution origin in the bed sediments of Aswan high dam reservoir:
lithogenic or anthropogenic sources**

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ABSTRACT

Heavy metals are present in aquatic sediments as a result of chemical leaching of bed rocks (Lithogenic source), and discharge of urban and industrial wastewaters (Anthropogenic sources). It is important to provide information about the source of heavy metals pollution because high levels of heavy metals can often be attributed to anthropogenic influences, rather than natural enrichment of the sediment by geological weathering (Lithogenic influences). This study was initiated with the objective of determining the origin of heavy metals pollutants in the bed sediments of Aswan High Dam Reservoir (AHD) and to evaluate their environmental risk in reservoir water and its potential use (benefits and risks) in agricultural land. Total metals concentration, the element Enrichment Factor (EF) and the sequential extractions of heavy metals (speciation) were determined in addition to pH, organic matter content and grain size distribution. The results indicated that the total metal contents varied in the following ranges (in mg/kg): Al (1165-18000), Cd (0.62-2.17), Co (11-47), Cr (12-63), Cu (80-261), Fe (3027-36651), Mn (93-1908), Ni (11-58), Pb (7-29) and Zn (98-208). The enrichment factor for all metals in sediment was found to be low and the heavy metals speciation revealed that the exchangeable and carbonate bound fractions are entirely insignificant. The results revealed no anthropogenic contribution of heavy metals and the chemical weathering is a primary factor that controls the concentrations of metals in the bed sediments of the Reservoir. The results indicated that metals in the bed sediments of the reservoir did not represent environmental pollution risk and are derived from lithogenic (natural) processes (i.e. weathering and soil formation). The bed sediments of the reservoir were found in the alkaline range and are promoting heavy metals precipitation. Thus it was recommended to use these sediments in soil amendment.

Keywords: Aswan High Dam Reservoir, Sediments, Enrichment Factor, speciation, lithogenic, anthropogenic.

INTRODUCTION

Aquatic sediments are principally derived from weathering processes, or from terrestrial sources under high runoff storms and floods. Heavy metals are present in aquatic sediments as a result of chemical leaching of bed rocks (Lithogenic source), and discharge of urban and industrial wastewaters (Anthropogenic sources). It is important to provide information about the source of heavy metals pollution because high levels of heavy metals can often be attributed to anthropogenic influences, rather than natural enrichment of the sediment by geological weathering (Lithogenic influences). Sediments from a habitat and energy supply for many freshwater organisms, which are used by man in food. The polluting substances are accumulated by sediments and then transferred to the aquatic organisms.

Sediments have been widely used as environmental indicators and their ability to trace contamination sources and monitor contaminants is largely recognized. They

play an important role in the assessment of metal contamination in natural waters (Wardas *et al.*, 1996). Pollution of the aquatic environment by heavy metals is a world-wide problem. Once heavy metals are discharged into water, they rapidly become associated with particulates and are incorporated in bottom sediments (Binning and Baird, 2001). Heavy metals such as cadmium, lead, copper and zinc are regarded as serious pollutants for aquatic ecosystems because of their environmental persistence, toxicity and ability to be incorporated into food chains (Förstner and Wittman, 1983). The accumulation of metals from the overlying water to the sediment is dependent on a number of external environmental factors such as pH, redox potential (Eh), ionic strength, anthropogenic input, the type and concentration of organic and inorganic ligands and the available surface area for adsorption caused by the variation in grain size distribution (Davies *et al.*, 1991). Heavy metal resulting from anthropogenic contamination are either associated with organic matter present in the thin fraction of the sediments, or adsorbed on Fe/Mn hydrous oxides, or precipitated as hydroxides, sulphides and carbonates (Förstner, 1985). Sediments represent one of the ultimate sinks for heavy metals discharged into the aquatic environment. Sediments act as both carriers and potential sources of contaminants in an aquatic environment. Increases in heavy metal concentrations tend to be associated with finer grained sediments, but the sandy character reduces their adsorptive properties, which cause a clear reduction of metal levels. Contaminants are not necessarily fixed permanently by the sediments, under changing environmental conditions, they may be released to water column by various processes of remobilization (Förstner and Wittmann, 1983). The occurrence of elevated levels of trace metals especially in the sediments can be a good indication of man-induced pollution where high levels of heavy metals can often be attributed to anthropogenic influences, rather than natural enrichment of the sediment by geological weathering (Davies *et al.*, 1991). It is important to distinguish between point source and diffuse inputs. The former includes effluent streams, drains discharges. Diffuse sources include aerial deposition and land runoff, particularly from rural areas. It has been widely recognized that identification of metal forms is necessary to understand their bioavailability and toxicity in the system (Fytianos, 2004). Use of total concentration as a criterion to assess the potential effects of the sediment contamination implies that all forms (phases) of a given trace metal have an equal impact on the environment. It is evident that just trace metal pollutants with the various sediment phases determine their specific impact on the environment. The total metal will only be able to provide information about the pollution if the background level is known, while metal origin (natural or anthropogenic) is rather difficult to predict. Thus to assess the environmental impact of sediments, determination of trace metals is not sufficient in itself (Salomons and Forstner, 1980). The chemical form (speciation) of the metal in the sediment ultimately determines behavior and mobilization ability of the metal in the environment. The term speciation of heavy metals in bottom sediments of natural and man-made water reservoirs refers to quantitative and qualitative differentiation of the forms of their occurrence. Therefore, this study was initiated with the objective of determining heavy metals pollution origin in the bed sediments of Aswan High Dam Reservoir and evaluate their environmental risk in the Reservoir water and its potential use (Benefits and Risks) in agricultural land, in addition to highlight relationships among metals and confirm the important role of the organic matter content and grain size on the distribution of metals in bottom sediments.

MATERIALS AND METHODS

Sampling

Superficial sediment samples were collected from ten sites along Aswan High Dam Reservoir during February 2007. The sampling sites were chosen to represent the different sedimentation areas (figure 1), where it started from Touthka (km 256.0) to El Daka (km 487.5), upstream the dam. The samples were transferred into labeled polyethylene bags and preserved frozen until they were analyzed in the laboratory.

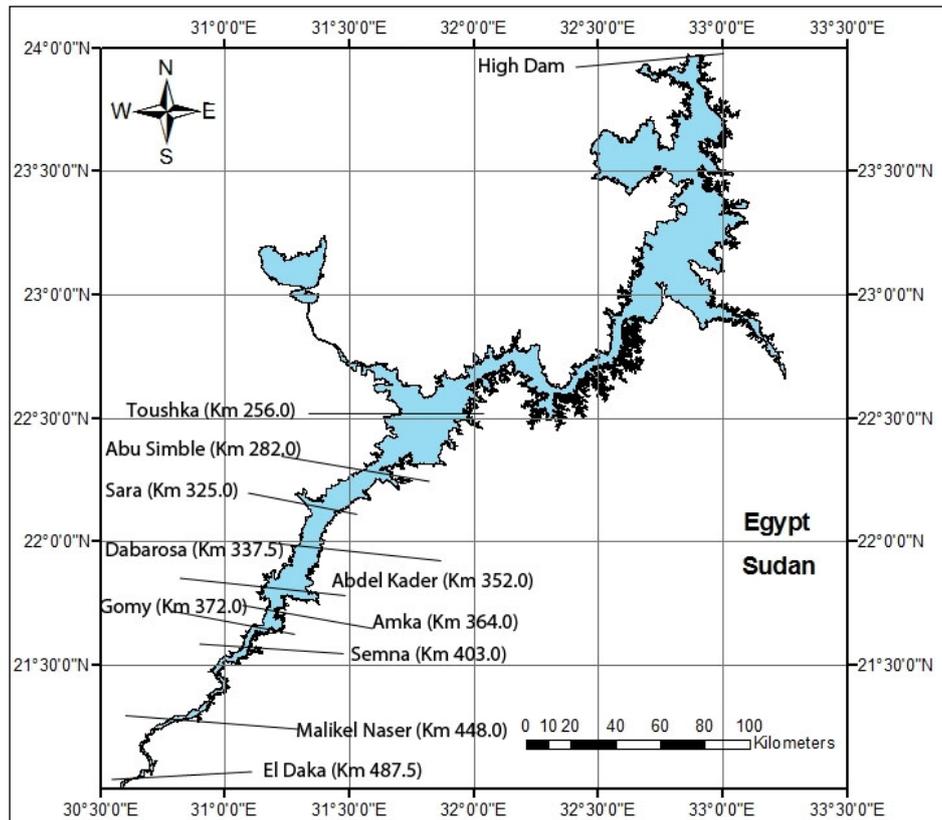


Fig. 1: Sampling sites along Aswan High Dam Reservoir

Sampling Analysis

During the analytical phase the particle size, the organic matter, pH and heavy metals were determined as follows:

Determination of Particle size

Particle size was performed on 50 g of sample that was dispersed using a dispersing agent and sonication for 10 min. The suspension was washed through <math><63\ \mu\text{m}</math> sieve to separate the sand fraction that was evaluated gravimetrically after drying. Coulter count measurements were carried out in the remaining suspension to determine silt and clay percentages (Santos *et al.*, 2003).

Determination of organic matter in bed sediments

Organic carbon is oxidized to carbon dioxide in the dichromate method with a parallel reduction of hexavalent chromium (Cr^{+6}) to trivalent chromium (Cr^{+3}) and an accompanying colour change from orange to green. The intensity of the green color was found to be proportional to the trivalent chromium concentration, which is related to the organic matter content (Howari and Banat, 2001).

Determination of heavy metals

Total heavy metals were determined using two grams of dry sample and mixed with 20 ml Aqua Regia (1:3 conc.HNO₃:conc.HCl) in a beaker and allowed to stand overnight. The mixture was heated to near dryness and allowed to cool then add 20 ml of 0.5 M HNO₃ acid solution. The samples were allowed to stand overnight then filtered (Kersten and Förstner, 1989; Binning and Baird, 2001). The total metals of the filtered solutions were measured using ICP instrument.

Sequential extraction in bed sediments

The sequential extraction procedures were carried out according to Tessier *et al.*, (1979) as follows:

Phase I (Exchangeable fraction): 3 g of the air-dried sediment sample is extracted with 20 ml 1.0M CH₃COONH₄, pH =7.0 for 30 min at room temperature and in agitation state.

Phase II (Bound to carbonates): the residue from the exchangeable fraction is extracted with 20 ml 1.0M CH₃COONa (adjusted to a pH of 5 with acetic acid) for 5 h at room temperature and in agitation state.

Phase III (Bound to Fe/Mn oxides): the residue from the carbonate fraction is extracted with 20 ml 0.04M NH₂OH.HCl (pH 2 with 25% acetic acid) for 85 overnight.

Phase IV (Organic and residual phases): which calculated from the difference between total metal content and the above three extracted phases.

Determination of pH

pH of the sediment was measured on a homogenized suspension of 10 g of sample with 25 ml distilled water after shaking for 10 min. after waiting 30 min, the measuring was carried out using pH meter WTW (Santos *et al.*, 2003).

Assessment of Heavy Metal Concentrations

Enrichment Factor (EF) was calculated according to Gonzalez *et al.*, (2000) to assess the heavy metal concentrations.

$$EF = (X_i)/(X_o) : (A_i)/(A_o)$$

where: (X_i); (A_i): concentrations of element X and reference element Al at sampling station i

(X_o); (A_o): background concentrations of element X and reference element Al.

Statistical Analysis

Correlation matrix was applied to study the behavior of the metal contents and their correlations in bed sediments using Minitab 12 Software.

RESULTS AND DISCUSSION

Characteristic of Sediments

The organic matter content, pH and particle size distribution of the sediments are presented in Table (1).

Grain size is important in sedimentation and transport processes. Sediments are characterized as coarse material, clay/silt and sand fractions, on the basis of separations, using 2 mm and 63 µm sieves. Particles >2 mm may consist of shells, rocks, wood, and other detrital materials, and are usually not a source of bioavailable contaminants (Mudroch *et al.* 1998). The clay/silt fraction has high surface area and because of its surface chemistry is more likely to adsorb organic and heavy metal contaminants. The percentages of clay and silt, along the reservoir ranged between 3.71%, at El Daka, and 99.87%, at Abu Simble. A high percentage of clay and silt in bottom sediment along Aswan High Dam Reservoir (99.87%) increases the ability of

sediments to adsorb the heavy metals and other pollutants. It is worth mentioning that, the higher concentration of heavy metals in the fine sediments is generally due to increase in specific surface area of clay minerals.

Table 1: Physicochemical characteristics of Aswan High Dam Reservoir sediments.

| Site Name | Km US AHD | Silt & Clay | Sand | pH | %OM |
|---------------|--------------|-------------|-------|------|------|
| Toushka | 256.0 | 99.52 | 0.48 | 7.59 | 2.1 |
| Abu Simble | 282.0 | 99.87 | 0.13 | 7.69 | 2.34 |
| Sara | 325.0 | 71.6 | 28.4 | 7.74 | 2.28 |
| Dabarosa | 337.5 | 98.18 | 1.82 | 7.92 | 1.05 |
| Abdel Kader | 352.0 | 97.84 | 2.16 | 7.7 | 2.36 |
| Amka | 364.0 | 97.8 | 2.2 | 7.54 | 2.97 |
| Gomy | 372.0 | 96.92 | 3.08 | 7.86 | 2.06 |
| Semna | 404.0 | 96.36 | 3.64 | 7.98 | 1.66 |
| Malikel Naser | 448.0 | 14.11 | 85.89 | 8.18 | 0.06 |
| ElDaka | 487.5 | 3.71 | 96.29 | 8.16 | 0.04 |
| Min | | 3.71 | 0.13 | 7.54 | 0.04 |
| Max | | 99.87 | 96.29 | 8.18 | 2.97 |

The pH values ranged between 7.54, at Amka, and 8.18, at El-Malk El Nasser. The low value of pH at Amka might be related to the high amount of organic matter. Generally, the pH values were found to be in the alkaline range, promoting heavy metals precipitation. Santos *et al.*, (2003) concluded that sediment pH is a decisive factor for heavy metal accumulation, where the pH increases the accumulation rate. This means that the use of these sediments in soil amendment is safe.

The organic matter contents along Aswan High Dam Reservoir, reach the highest value at Amka (2.97%). The lowest value of organic matter was recorded at El-Daka (0.04%). The role of organic matter and sediment grain size in relation to the accumulation of heavy metal to the sediment has been emphasized by Davies *et al.*, (1991). In general, organic matter and clay texture play an important role in controlling heavy metal concentrations in bottom sediments. Gaiero *et al.* (1997) stated that the organic matter exhibited a closer correlation with metals probably caused by a larger capacity for complexation in the biologically processed organic debris. The behavior of natural organic matter might be the most important factor that influences the trace metal cycling in aquatic systems (Barry, 1982).

Total Heavy Metal Concentrations

The total heavy metal concentrations in bed sediments at different sampling sites were illustrated in figure (2). The results showed that total metal contents varied in the following ranges (in mg/kg): Al (1165-18000), Cd (0.62-2.17), Co (11-47), Cr (12-63), Cu (80-261), Fe (3027-36651), Mn (93-1908), Ni (11-58), Pb (7-29) and Zn (98-208). The metal concentrations decreased in the order Fe > Al > Mn > Cu > Zn > Cr > Ni > Co > Pb > Cd. The results showed that the highest contents of Fe, Zn and Co were observed in Amka with the values of 36651, 208 and 47 mg/kg, respectively. The concentrations of Cd, Cr and Mn in Gomy site were higher than those obtained from other sites. In addition, the highest concentrations of Pb and Ni were also high in Abu-Simble with values of 29 and 58 mg/kg, respectively. The metal concentrations in El-Daka site were found significantly lower than those found in other sites.

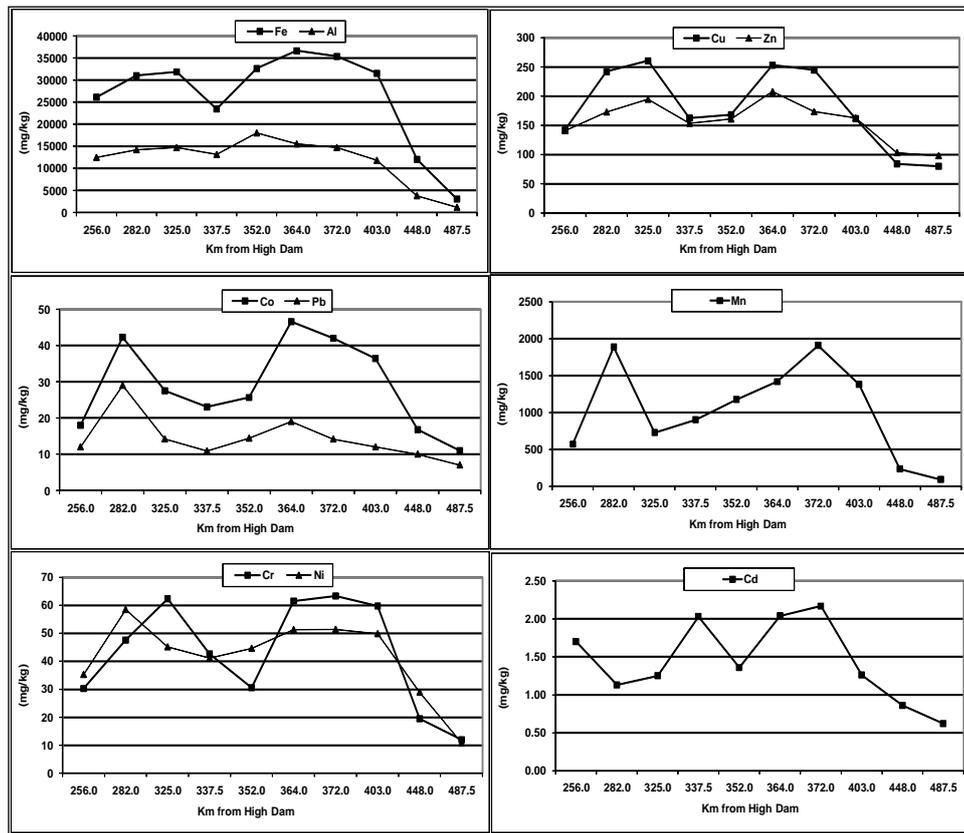


Fig.2: Heavy metals concentration in bed sediment of Aswan High Dam Reservoir.

Correlation Matrix

Correlation matrix between different variables (Table 3), indicated a significant positive correlation among the concentration of metals along Aswan High Dam Reservoir ($P < 0.05$). Heavy metals in general have positive coefficients and mostly are significantly correlated. Organic matter exhibited a high degree of selectivity and correlation with some heavy metals (Davies, 1984).

Table 3: Correlation matrix between different physico-chemical parameters

| | Al | Cd | Co | Cr | Cu | Fe | Mn | Ni | Pb | Zn | OM | pH | Silt and Clay | Sand |
|---------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|---------------|-------|
| Al | | 0.67 | 0.66 | 0.69 | 0.79 | 0.94 | 0.72 | 0.85 | 0.54 | 0.86 | 0.91 | -0.82 | 0.92 | -0.92 |
| Cd | 0.67 | | 0.53 | 0.60 | 0.56 | 0.67 | 0.54 | 0.55 | 0.16 | 0.62 | 0.57 | -0.56 | 0.74 | -0.74 |
| Co | 0.66 | 0.53 | | 0.85 | 0.84 | 0.82 | 0.92 | 0.88 | 0.73 | 0.83 | 0.75 | -0.54 | 0.66 | -0.66 |
| Cr | 0.69 | 0.60 | 0.85 | | 0.88 | 0.85 | 0.75 | 0.83 | 0.46 | 0.90 | 0.71 | -0.49 | 0.68 | -0.68 |
| Cu | 0.79 | 0.56 | 0.84 | 0.88 | | 0.85 | 0.76 | 0.83 | 0.69 | 0.95 | 0.85 | -0.70 | 0.68 | -0.68 |
| Fe | 0.94 | 0.67 | 0.82 | 0.85 | 0.85 | | 0.82 | 0.93 | 0.57 | 0.92 | 0.93 | -0.77 | 0.89 | -0.89 |
| Mn | 0.72 | 0.54 | 0.92 | 0.75 | 0.76 | 0.82 | | 0.90 | 0.72 | 0.72 | 0.71 | -0.50 | 0.76 | -0.76 |
| Ni | 0.85 | 0.55 | 0.88 | 0.83 | 0.83 | 0.93 | 0.90 | | 0.75 | 0.85 | 0.81 | -0.63 | 0.84 | -0.84 |
| Pb | 0.54 | 0.16 | 0.73 | 0.46 | 0.69 | 0.57 | 0.72 | 0.75 | | 0.60 | 0.65 | -0.60 | 0.52 | -0.52 |
| Zn | 0.86 | 0.62 | 0.83 | 0.90 | 0.95 | 0.92 | 0.72 | 0.85 | 0.60 | | 0.90 | -0.76 | 0.76 | -0.76 |
| OM | 0.91 | 0.57 | 0.75 | 0.71 | 0.85 | 0.93 | 0.71 | 0.81 | 0.65 | 0.90 | | -0.93 | 0.83 | -0.83 |
| pH | -0.82 | -0.56 | -0.54 | -0.49 | -0.70 | -0.77 | -0.50 | -0.63 | -0.60 | -0.76 | -0.93 | | -0.77 | 0.77 |
| Silt and Clay | 0.92 | 0.74 | 0.66 | 0.68 | 0.68 | 0.89 | 0.76 | 0.84 | 0.52 | 0.76 | 0.83 | -0.77 | | -1.00 |
| Sand | -0.92 | -0.74 | -0.66 | -0.68 | -0.68 | -0.89 | -0.76 | -0.84 | -0.52 | -0.76 | -0.83 | 0.77 | -1.00 | |

The organic matter play an important role in controlling heavy metal concentrations in the bottom sediments, indicating that anthropogenic metals readily allow them to substitute in the carbonate, organic matter and sulfide phases. The high correlation values between the organic content and the heavy metal is related to the considerable chelating power between them as well as to the power of the surface adsorption of the heavy metals onto the surface of the organic material (Gupta and Subramanian, 1998). Iron showed closer association with most heavy metals along the

reservoir. A significant correlation between iron and most metals indicated co-precipitation of these metals with iron hydroxides. Worthy of attention is also the fact that all metals are positively correlated with organic matter ($p < 0.05$). The strong associations between the heavy metals suggest their possible common sources from heavy minerals and / or chemical similarity. It is worth to mention that there is a highly negative correlation between pH and organic matter.

Metal Pollution Assessment

The assessment of heavy metals in this study was carried out with the objective of determining heavy metals pollution origin in the bed sediments of Aswan High Dam Reservoir and evaluate their environmental risk in reservoir water and its potential use (benefits and risks) in agricultural land. Therefore, enrichment factor (EFs) and metal speciation were applied.

Enrichment Factors (EFs)

The background metal concentration used in the calculation of Enrichment Factors (EFs) were selected in this study as the reference point taken from El-Daka site (km 487.5). The results of the calculation of the Enrichment Factors (EFs) for the metal under investigation relative to the content of Al (reference element) at each site are presented in Figures (3). Assuming an element concentration higher than twice the background content implies anthropogenic pollution, EFs higher than 2 indicate contaminated sites (Sutherland, 2000).

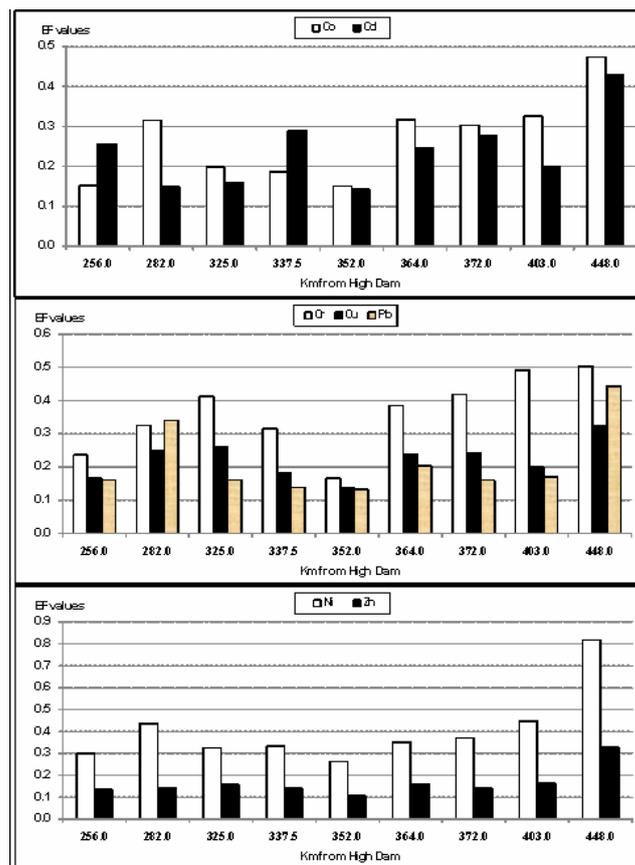


Fig. 3: Enrichment Factor for different heavy metals in bed sediment of AHD Reservoir

Based on this classification the bed sediments along Aswan High Dam Reservoir are not contaminated by metals analyzed in this study (EFs less than 2) suggesting that no anthropogenic contribution of heavy metals and the chemical

weathering is the primary factor that control the concentrations of metals in Aswan High Dam Reservoir sediments.

Metal Speciation

Metal speciation results can provide information on heavy metals pollution origin in the bed sediments of Aswan High Dam Reservoir and possible chemical forms of heavy metals in sediments to evaluate their environmental risk in reservoir water.

Figure (4) presented the results of heavy metal speciation in bed sediment along Aswan High Dam Reservoir. These results showed that, High percentages of heavy metals were found in phase IV (organic and residual) which ranged between 74.29% for Zn and 91.83% for Cu, whereas much lower percentages were found in phase I (exchangeable) and phase II (carbonate). These two phases are the most easily released to water and are considered as bioavailable. Only minor amounts of metals were detected in the exchangeable phase (0.03% - 2.36%). The contribution of the phase II (bound to carbonates) was very small (0.39% - 4.75%). It is worth to mention that, the metals occurring in phase I and II can be released to bulk water as a result of change in the ionic composition of water or a decrease in water pH.

Phase III (Metals bound to iron and manganese oxides) is sensitive to the redox potential changes and in anaerobic conditions become unstable. The contributions of chromium and zinc in the form of phase III were found higher than the contributions of other metals (16.86% and 18.6%, respectively). This result agrees with the high correlation found between iron and both chromium ($r=0.85$) and zinc ($r=0.92$), that confirm the association of Cr and Zn with amorphous oxides. Metal Speciation results revealed that the exchangeable and carbonate bound fractions are entirely insignificant (negligible pollutants). This result revealed that no anthropogenic contribution of heavy metals and the chemical weathering is the primary factor controlling the concentrations of metals in the bed sediments of the reservoir.

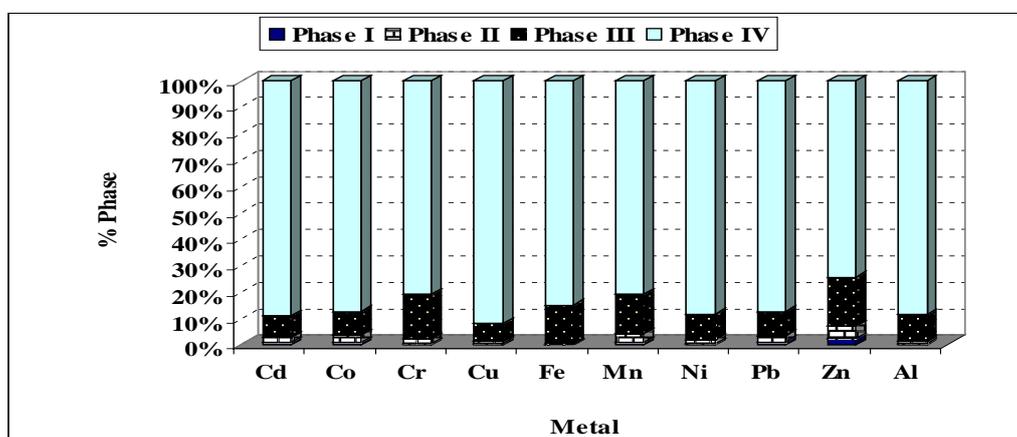


Fig. 4: Metals speciation in bed sediment of Aswan High Dam Reservoir

CONCLUSIONS

From the above results, the following conclusions can be reached:

- The heavy metal concentrations in bed sediments at different sampling sites are decreased in the order $Fe > Al > Mn > Cu > Zn > Cr > Ni > Co > Pb > Cd$ according to their concentrations.
- Calculations of the enrichment factor for all metals and their speciation analyses revealed that the heavy metals in the reservoir are negligible pollutants, and

should be considered as a natural background; mainly derived from lithogenic processes such as weathering and soil formation.

- The bed sediments of the reservoir were found in the alkaline range, promoting heavy metals precipitation. So, the use of these sediments in soil amendment is safe.
- It is recommended to implement a pilot project to evaluate the characteristics of sediment for its potential use in agricultural land reclamation.

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ARABIC SUMMERY

مصدر التلوث بالعناصر الثقيلة في رسوبيات خزان السد العالي بأسوان: مصادر طبيعية أو بشرية

محسن يسري - طلعت سالم - كريمة محمود
معهد بحوث النيل- المركز القومي لبحوث المياه

المعادن الثقيلة موجودة في الرواسب المائية نتيجة لتسرب المواد الكيميائية للصخور (مصدر طبيعي)، وتصريف مياه الصرف الصحي في المناطق الحضرية والصناعية (المصادر البشرية). ومن المهم توفر معلومات حول مصدر التلوث بالمعادن الثقيلة لأن مستويات عالية من المعادن الثقيلة يمكن في كثير من الأحيان أن تعزى إلى التأثيرات البشرية، بدلا من الطبيعية التي تنتج من الرواسب الجيولوجية بواسطة التجوية. وقد بدأت هذه الدراسة بهدف تحديد منشأ الملوثات بالمعادن الثقيلة في رواسب قاع خزان السد العالي بأسوان (AHD) وتقييم مخاطرها البيئية في مياه الخزان واستخدام إمكاناتها (الفوائد والمخاطر) في الأراضي الزراعية. وقد تم تحديد مجموع تركيز المعادن، وعامل إثراء العنصر (EF) والأستخلاص بالتتابع للمعادن الثقيلة بالإضافة إلى درجة الحموضة ومحتوى المادة العضوية وتوزيع حجم الحبيبات. وقد دلت النتائج على أن مجموع محتويات العناصر الكلية يقع في النطاقات التالية (ملجم / كجم): الألومنيوم (١١٦٥ - ١٨٠٠٠)، الكاديوم (٠,٦٢ - ٢,١٧)، الكوبلت (١١-٤٧)، والكروم (١٢-٦٣)، النحاس (٨٠-٢٦١)، الحديد (٣٠٢٧ - ٣٦٦٥١)، والمنجنيز (٩٣ - ١٩٠٨)، النيكل (١١-٥٨)، والرصاص (٧-٢٩) والزنك (٩٨ - ٢٠٨). ولقد وجد أن معامل الإثراء لجميع المعادن في الرسوبيات منخفض كما أن فصل العناصر الثقيلة بالتتابع بين أن كلا من الصورة التبادلية والتي علي هيئة الكربونات ضئيلة تماما. وكشفت النتائج أنه لا توجد مساهمة بشرية من المعادن الثقيلة وأن التجوية الكيميائية هي العامل الأساسي الذي يتحكم في تركيزات المعادن في رواسب قاع الخزان. كما أشارت النتائج إلى أن المعادن في رواسب قاع الخزان لا تمثل مخاطر للتلوث البيئي وأنها مستمدة من الطبيعية نتيجة عمليات التجوية وتكوين التربة. وقد وجد أن رسوبيات قاع الخزان تقع في حدود القلوية مما يعزز ترسيب المعادن الثقيلة. لذلك نوصي باستخدام هذه الرسوبيات في تحسين التربة.