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Spatial distribution and pollution assessment of trace metals in surface sediments of Ziqlab Reservoir, Jordan

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Abstract Surface sediment samples were collected from Ziqlab dam in northwestern Jordan to investigate the spatial distribution of selected trace metals and assess their pollution levels. The results showed that the concentrations of Pb, Cd, and Zn exceeded the environmental background values. Cd, Ni, and Cr contents were higher than the threshold effect level (TEL) in 63, 83, and 60 % of the reservoir sediments, respectively; whereas Pb, Zn, and Cu were less than the TEL limit. The concentrations of trace metals in reservoir sediment varied spatially, but their variations showed similar trends. Elevated levels of metals observed in the western part (adjacent to the dam wall) were coincided with higher contents of clay-silt fraction and total organic matters. Multivariate analysis indicated that Pb, Co, and Mn may be related to the lithologic component and/or the application of agrochemicals in the upstream agricultural farms. However, Cd and Zn concentrations were probably elevated due to inputs from agricultural sources, including fertilizers. Evaluation of contamination levels by the Sediment Quality Guidelines of the US-EPA, revealed that sediments were non-polluted to moderately polluted with Pb, Cu, Zn, and Cr, but non-polluted to moderately to heavily polluted with Ni and non-polluted with Mn. The geoaccumulation index showed that Ziqlab sediments were unpolluted with Pb, Cu, Zn, Ni, Cr, Co, and Mn, but unpolluted to moderately polluted with Cd. The high enrichment values for Cd and Pb (>2) indicate their anthropogenic sources, whereas the remaining elements were of natural origins consistent with their low enrichment levels.

Keywords Metal contamination · Reservoir sediments · Sediment quality guidelines · Geoaccumulation index · Enrichment factor · Jordan

Introduction

Jordan faces growing water challenges and acute water shortage crisis seems inevitable. Even with more sustainable practices, the future of water resources in Jordan is uncertain. Rapidly growing populations, economic development, increasing standards of living, and competing demands for water resources all put strains on scarce resources (Drake 1997; Al-Rawabdeh et al. 2013; Al-Taani et al. 2012; Al-Taani 2013, 2014). In

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addition to limited water supply, extreme and prolonged droughts have already exacerbated the water shortage problems.

The growing debate over the water issues, which has lately been intensified with the recent influxes of Syrian refugees, has forced the government to respond quickly. A number of short- and long-term alternatives were explored to increase Jordan's water supply, of which dam building remain a viable (but costly) solution. While water reserves will not solve the country's water problems, they will reduce Jordan's water deficit, particularly in the agricultural sector. Agriculture is the largest user of water with 72 % of total water demand in 2005 (MWI 2008), though the agricultural sector contributed only 2.7 % of Jordanian GDP in 2004 (Dean 2005).

Despite the increasing reliance on water dams, there is a general decrease in rainfall totals (due to climate change), where flash floods become more common. Flash floods can accelerate weathering of rocks and increase the quantities of sediments (and pollutants) discharged into the dams. High-intensity rainfall generally mobilizes more nutrient-laden sediment than low-intensity storm events (Basnyat et al. 2000). In addition, water reservoirs are vulnerable to pollution from various human activities.

Among others, trace metals are one of the major groups of contaminants. Metals are widespread and naturally occurring in all environments. While some are nutritionally essential (in low concentrations), others are potentially toxic. In addition to being toxic to humans, the heavy metals (e.g., mercury, lead, and cadmium) in water pose a potential threat to aquatic ecosystems if present in high concentrations due to their toxicity, persistence, and bioaccumulation characteristics (Pekey 2006; Osher at al. 2006; Lafabrie et al. 2007; Diagomanlin et al. 2004; Demirak et al. 2006; Zheng et al. 2008; Batayneh 2010).

Metals in aquatic environments are largely accumulated in suspended particles and sediments, where the sediments act as a sink and a source of the metals. The assessment of trace metal distribution in sediments can provide clues about the magnitude of human activities and their effects on ecosystems. In addition, they can help evaluate the risks associated with discharged human waste. The contamination of surface sediments with heavy metals can have an impact on water quality and the organisms that make up the base of the food chain, ultimately affecting human health (Christophoridis et al. 2009).

The input of metal contaminants can be of geogenic or anthropogenic sources (Wittmann et al. 1981; Fan 1996; He et al. 2005; Bai et al. 2011a, b, c; Al-Taani et al. 2012, 2013, 2014; Batayneh 2012; Batayneh et al. 2012, 2014; Sakan et al. 2012; Gao et al. 2013). The pathways and rates of mobilization of metals depend primarily on the physico-chemical conditions of water and chemical characteristics of the metals.

Ziqlab dam, in northwest Jordan, was built in 1966 in response to the growing water demands. However, the high rate of sedimentation and subsequent release of trace metals has affected the quality of water and became a growing concern (El-Radaideh et al. 2014). This study evaluates the levels and spatial distribution of trace metals in the surface sediments of Ziqlab dam. It also assesses the potential sources of trace metal and pollution levels in sediments using the Sediment Quality Guidelines of the US-EPA, the geoaccumulation index, and enrichment factor methods. This evaluation helps develop effective measures for protecting Ziqlab reservoir's water and better management of human activities in the dam's watershed.

Materials and methods

Study area

Ziqlab dam, which was constructed in 1966, is located in northwestern Jordan (Fig. 1). The dam was proposed in response to the growing water demands, particularly for irrigation. The reservoir is 1500 m in approximate length with a width ranging between 35 and 500 m and a storage capacity of 4.4 million cubic meters (MCM) (Shatnawi 2002).

The Ziqlab watershed is about 106 km^2 and is characterized by a semi-humid (in the east) to arid climate (in the western part). The Mesozoic deposits, mainly chalk, marl, limestone, phosphatic limestone, and chert, cover the majority of the Ziqlab catchment area. The Cenozoic rocks contain basaltic lava, conglomerate, silt, clay, and gypsum (Abed and Ashour 1987). Unconsolidated alluvial sediments and alluvial fans are spread over the Jordan River forming an aquifer in the northwest area of Jordan. The southern Ziqlab watershed is composed of fluvial, alluvial, and organic deposits. The lower portion of Ziqlab catchment contains outwash materials consist of rounded gravels mixed with sand and occasionally with minor amounts of silt.

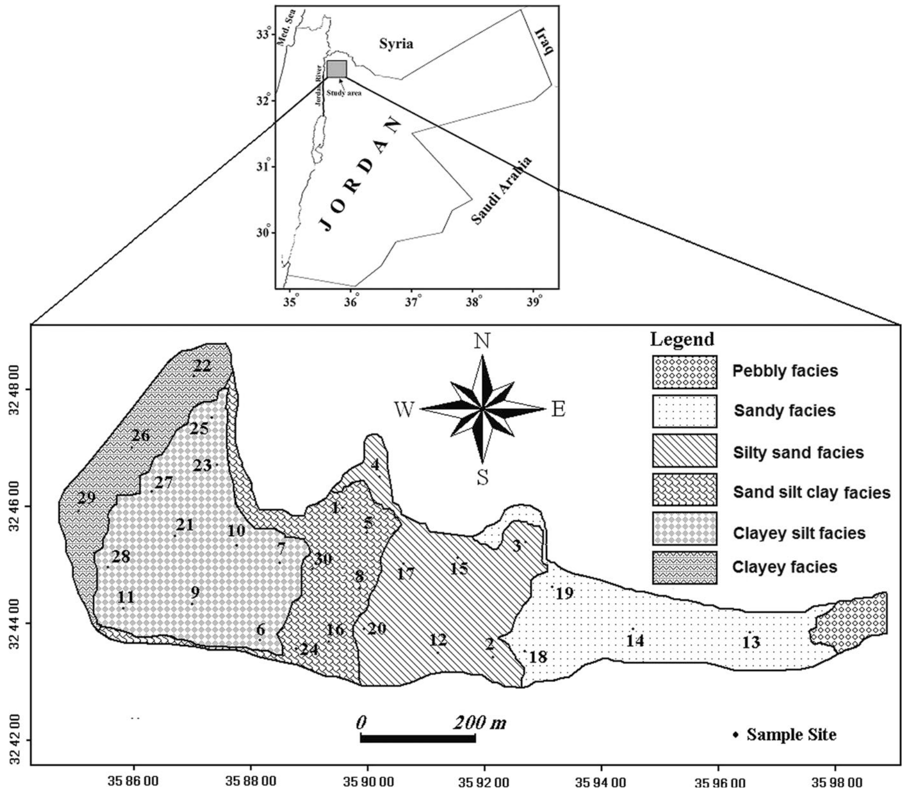

Fig. 1 Site map of Ziqlab reservoir including sampling sites and sediment facies map

The volume of accumulated sediment was estimated to be greater than 2 MCM (El-Radaideh et al. 2014), with an annual sediment accumulation rate of about 0.046 MCM. The sources of sediments in the reservoir are diverse, and include assemblages of secondary minerals. Mineral or organic nutrients are concentrated in places where clay and silt are the major sediment types found at the bottom of the Ziqlab reservoir.

Analytical methods

Thirty surficial sediment samples (from 1 to 30 cm deep) were collected from Ziqlab reservoir in May 2012 using a Van Veen grab sampler. Sample preparation and handling were done according to 1981 US Environmental Protection Agency methods (EPA/CE-81-1 Protocol). Sediments were initially characterized for grain size distribution. Sediment samples were then digested using a mixture of $HClO_4$, HNO_3, and HF in Teflon tubes for analysis of the total concentrations of Fe, Pb, Cd, Cu, Zn, Ni, Co, Cr, and Mn. The digested sample solutions were analyzed using Atomic Absorption Spectroscopy (Philips SP 9PYE Unicam Spectrophotometer). Quality assurance and quality control were assessed using duplicates and method blanks. A standard sediment reference material (BCSS-1), issued by the State Oceanographic Administration of China, was subjected to the same digestion technique and was run concurrently along with the samples (Table 1). Average recoveries of Pb, Cd, Cu, Zn, Ni, Co, Cr, and Mn in triplicate analysis were 103±4.0, 94±2.0, 95±3.2, 93±6.1, 97±3.7, 97±4.2, 98±2.9, and 98±4.0 %, respectively (Table 1). Analytical precision was <10 % for all of the elements. The cation exchange capacity (CEC) was determined by measuring the sodium content after treatment of 5 g, taken from sediment fraction of less than 63 μm, with sodium acetate and ammonium acetate solution (Loring and Rantala 1992; Hesse 1972). Total organic matter (TOM) was measured with the titration method (Hesse 1972). Soil pH was measured using a Hach pH meter (Hach Company, Loveland, CO, USA) (soil/water=1:5).

Table 1 Comparison of BCSS-1 certified values for total trace metals (in mg kg^{-1})

Elements	Present study	BCSS-1	Recovery percent
Pb	23.4	22.7	103 (±4)
Cd	0.28	0.3	94 (±2)
Cu	17.5	18.5	95 (±3.2)
Zn	110.4	119	93 (±6.1)
Ni	53.4	55.3	97 (±3.7)
Co	11.09	11.4	97 (±4.2)
Cr	120.8	123	98 (±2.9)
Mn	224.2	229	98 (±4)

Statistical analysis

Data analysis was carried out using SPSS 16.0 for Windows (SPSS Munchen, Germany). Correlation analysis (Pearson's method) was performed to identify the relationships between the concentrations of trace elements and sediment properties. Hierarchical cluster analysis (HCA) was applied to a subgroup of the hydrogeochemical dataset. Principal component analysis (PCA) also used to determine the common pollution sources. The PCA was applied as the frequency diagrams of chemical parameters do not follow a normal distribution. Therefore, PCA was carried out for the logarithmic transformation of data set because it is closer to the normality condition which is required for these analyses. Standardization was applied to the log normal distribution to ensure that each variable was weighted equally. Log-transformation of positively skewed chemical parameters as well as data standardization is commonly done in multivariate statistical analysis.

PCA reduces a large number of variables in to a smaller number, and for further investigation of the relationships between the elements. The principal components (PCs) with eigenvalues larger than 1 are extracted with the PC loadings rotated for the maximum variance. A total of two PCs are extracted, which account for 87.75 % of the total variance.

Results and discussion

Physico-chemical properties of sediments

The grain size analysis of the surface sediments of Ziqlab reservoir is tabulated in Table 2. Sediments are largely composed of sand, silt, and clay, deposited in the old watercourse bed in the reservoir, with an average grain size that increases toward the reservoir shores (Fig. 1). Table 2 also shows that the TOM content in reservoir sediments ranges between 1.7 and 8.5 %, with an average of 5.4 %. High TOM values were observed in clay-silt facies but the values decreased in sandy facies (<2 %; Table 2, Figs. 1 and 2). This is probably related to the high energy level of the reservoir, which suggests a higher input of allochthonous organic matter into the system (El-Radaideh et al. 2014).

The TOM is probably attributed to the organic content leached from bituminous limestone, oil shale, phosphate, and biogenic phytoherm tufa deposits exposed in the catchment. Furthermore, floodwaters wash out untreated wastewater from adjacent towns along Ziqlab Wadi into the reservoir (Saadoun et al. 2008).

The pH of the reservoir sediments varies from 5.6 to 7.8, with an average of 6.8 (Table 2). These values are relatively suitable for metal release from sediment, where the lower the pH, the higher the metals released. The CEC levels in the reservoir sediment range between 65 and 160 meq/100 g, with a mean value of 105 meq/100 g (Table 2). The high CEC in the clay and silt is probably related to the high content of available clay minerals (Spisto 1989; El-Radaideh et al. 2014).

Trace element levels

The statistical analysis of trace element concentrations in the surface sediments of Ziqlab reservoir is summarized in Table 3. The majority of metals are moderately variable, with the highest spatial variability observed for Cd (with coefficient of variation of 0.71). According to Taylor (1964) and He et al. (2005), the average concentrations of Pb, Cd, and Zn exceed the background levels as they were approximately 2.08, 3.8, and 1.04 times higher than their respective background values (Table 3). Based on the sediment guidelines for freshwater ecosystems (Smith et al. 1996; MacDonald et al. 2000; Caeiro et al. 2005), the mean levels of Cd, Ni, and Cr are higher than the threshold effect level (TEL) in 63, 83, and 60 % of bottom sediment of Ziqlab Reservoir, respectively. The TEL is assumed to represent the concentration below which toxic biological effects rarely occur. In the range of concentrations between the TEL and probable effect level (PEL), toxic effects occasionally occur, at concentrations above the PEL, toxic effects usually or frequently occur (USEPA 1997). However, the average

Table 2 Textural and chemical data for surficial sediments (1–30 cm deep) of Ziqlab reservoir, Jordan

Sample no.	Textured facies	Sand %	Silt	Clay	TOM	pH	CEC meq/100 g
1	Sand-silt-clay	40	37	23	3.7	6.8	100
2	Silty-sand	58	27	14	4	6.6	100
3	Silty-sand	68	21	10	4.2	6.9	95
4	Silty-sand	67	22	11	3.6	6.5	90
5	Sand-silt-clay	31	43	25	4.6	7	115
6	Clayey-silt	3.7	60	36	7.2	7.1	110
7	Clayey-silt	5.7	58	37	7.6	7.3	105
8	Clayey-silt	4.9	60	35	7	7.5	100
9	Sand-silt-clay	21	50	29	6	6	120
10	Clayey-silt	7	59	35	6.9	6.6	110
11	Clayey-silt	15	55	30	6.8	6.7	120
12	Silty-sand	53	36	10	3.9	6.9	90
13	Sand	76	18	6	1.9	5.6	80
14	Sand	97	2	1	2	6	75
15	Silty-sand	55	33	11	3.5	6.5	110
16	Sand-silt-clay	27	51	22	7	6.8	90
17	Silty-sand	49	27	14	3.5	7.2	100
18	Sand	79	13	7	1.7	6.1	70
19	Sand	90	6	4	1.8	6.3	65
20	Silty-sand	53	35	13	2.9	7.5	80
21	Cayey-silt	2.6	57	40	7.2	6.9	120
22	Clay	1	9	90	8.5	7.6	140
23	Cayey-silt	8	56	57	7.1	6.9	130
24	Sand-silt-clay	37	39	24	5.5	6.9	100
25	Cayey-silt	1	57	42	7.1	6.8	110
26	Clay	2	7	91	8	7.1	130
27	Clayey-silt	7	62	31	6.9	6	120
28	Clayey-silt	8.9	67	23	7.2	6.3	125
29	Clay	7	3	90	7.9	7.8	160
30	Sand-silt-clay	27	51	22	6.9	6.5	100
Mean		33	37	29	5.4	6.8	105.3
Min.		1	2	1	1.7	5.6	65
Max.		97	67	91	8.5	7.8	160
St. Dev.		30	21	24	2.2	0.5	21.09

SD standard deviation

concentrations of Pb, Zn, and Cu are less than the TEL. Nevertheless, all metals are below the PEL (Table 3). Cd, Ni, and Cr levels exceed the TEL limit, thus posing serious pollution threats. Ziqlab bottom sediments appear to be slightly polluted with Pb, Zn, and Cu, as about 70–96 % of reservoir sediments are below the TEL limit.

Spatial distribution of trace elements

Spatial distribution patterns of Pb, Cd, Cu, Zn, Ni, Co, Cr, and Mn in the surface sediments of Ziqlab Reservoir are shown in Fig. 2. Higher levels of Pb are located in the west (dam's outlet) (Fig. 3a), whereas lower values are found in the eastern corner of the reservoir adjacent

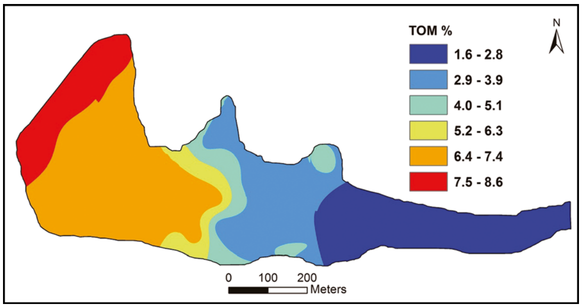

Fig. 2 Spatial distribution patterns of TOM contents in surface sediments of Ziqlab dam

to dam's water inlet. These spatial distribution trends are closely linked to grain size distribution (Fig. 1), where clay-silt facies are dominant in the western side, while highly drained and coarse facies are accumulated in the eastern reservoir. Highly positive correlations were found between Pb and TOM, CEC, clay contents with r=0.89, 0.74, and 0.73, respectively, but the correlation coefficient becomes moderate between Pb and pH (r=0.51) (Table 4). The distribution pattern of Cd (Fig. 3b) was similar to that of Pb, with positively significant correlation (r=0.79) observed between Cd and Pb (Table 4). Similarly, highly positive correlations (p<0.01) were found between Cd and clay, TOM and CEC (Table 4).

Likewise, the concentrations of Cu, Zn, Ni, Co, Cr, and Mn varied spatially with values that increased in the western direction, where the predominant fine fraction occurred, and decreased eastwards closer to the dam's inlet, the portion with high percent of coarser grain-sized sediments (Figs. 3c–h) and 1). These metals were also positively correlated (p<0.01) with clay, pH, TOM, and CEC levels (Table 4).

The similar spatial variations of these trace elements in the bottom sediment of the reservoir are closely related to the similar geological enrichment characteristics, and probably indicate similar sources (Bai et al. 2011a; El-Radaideh et al. 2014). In addition, these spatial trends of trace metal concentrations may be attributed to TOM contents (2–9 %, Table 2 and Fig. 2) which also exhibited similar spatial variations. This is consistent with significantly positive correlations found between trace metals and TOM levels (Table 4). This can be explained by the significance role of organic matter to the binding of certain heavy metals (Shea 1988; Murray et al. 1999; Eimers et al. 2002). Kalbitz and Wennrich (1998) and Gonzalez et al. (2006) reported that organic matter could act as a major sink for trace elements due to its strong ability to form complexes with metallic contaminants. The role of organic matter in accumulating trace metals to sediments was also emphasized by Wakida et al. (2008).

Table 3 Summary statistics of trace element content in surficial sediments of Ziqlab reservoir, compared to other reference concentrations

Sediment samples (n=30)	Fe %	Pb	Cd	Zn	Ni mg kg^{-1}	Co	Cr	Cu	Mn
Minimum	1.1	2	0.1	10	13	2	3	4	11
Maximum	2.9	49	1.9	120	88	18	65	31	100
Average	2.03	26	0.76	72.9	45.4	9.9	38.6	16.6	58.1
S.D.	0.36	12.6	0.54	34.9	21.9	3.5	18.7	7.4	26.3
CV	0.18	0.49	0.71	0.48	0.48	0.4	0.49	0.44	0.45
Background[a]	3.8	12.5	0.2	70	75	25	100	55	950
TEL[b]	–	35	0.6	123	22.7	–	37.3	35.7	–
PEL[b]	–	91.3	3.53	315	48.6	–	90	197	–
SQG[c]									
Non-polluted	–	<40	–	<90	<20	–	<25	<25	<300
Moderately polluted	–	40–60	–	90–200	20–50	–	25–75	25–50	300–500
Heavily polluted	–	>60	>6	>200	>50	–	>75	>50	>500

SD standard deviation, *CV* coefficient of variation, *TEL* threshold effect level, *PEL* probable effect level

[a] From Taylor (1964) to He et al. (2005)

[b] From Smith et al. (1996), MacDonald et al. (2000) to Caeiro et al. (2005)

[c] From Giesy and Hoke (1990)

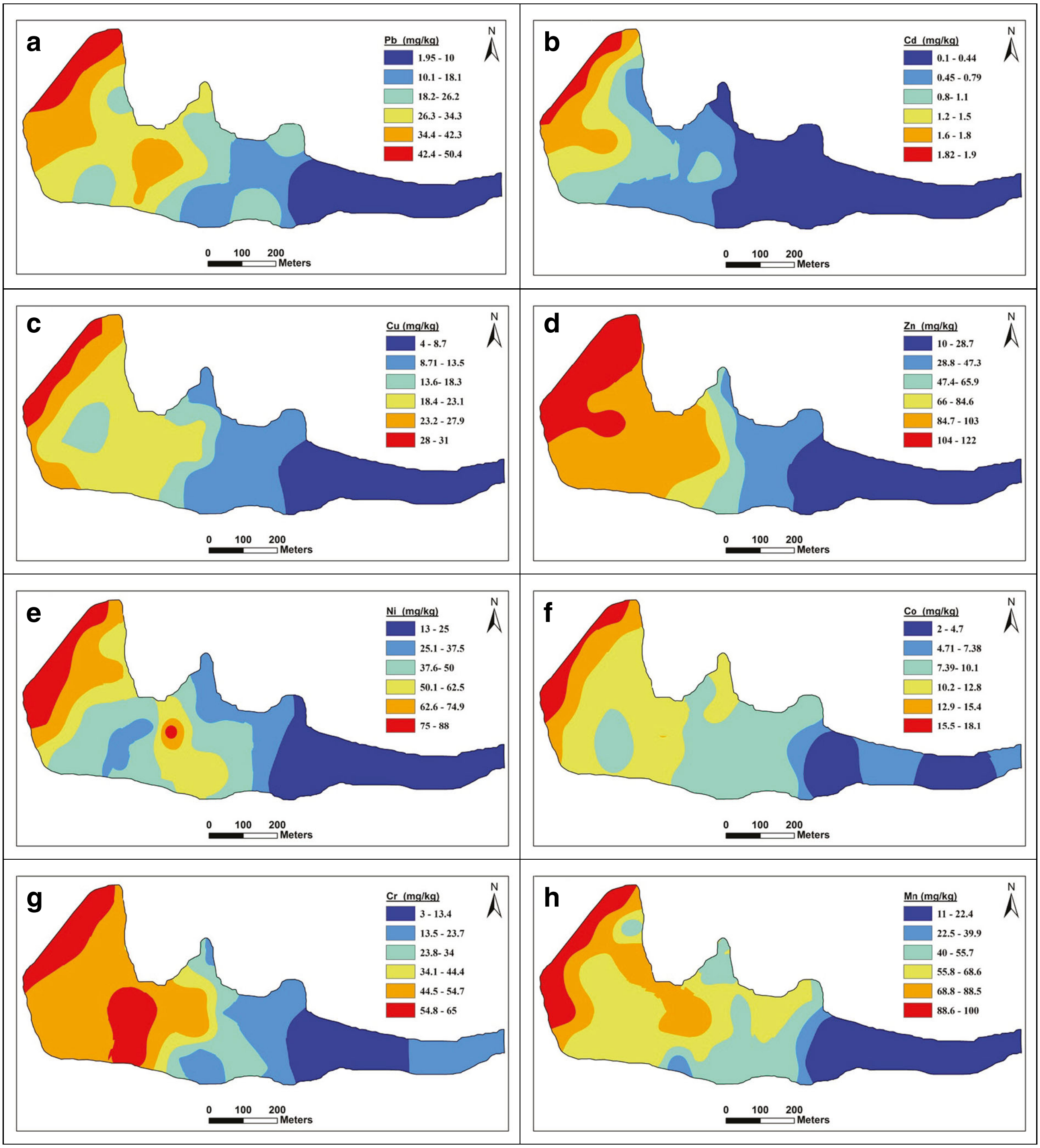

Fig. 3 Spatial distribution patterns of trace metal contents **a** Pb, **b** Cd, **c** Cu, **d** Zn, **e** Ni, **f** Co, **g** Cr, and **h** Mn in surface sediments of Ziqlab dam

HCA and PCA

The dendrogram in Fig. 4 shows the concentrations of the eight elements (using HCA analyses) deduced from the 30 sediment samples. The elements are classified into two different groups: C1 and C2 (Fig. 4). Group C1 (Zn, Cr, Cu, Pb, and Cd) has lower linkage distances, but greater similarity, compared to group C2 (Co, Mn, and Ni). The lowest linkage distances observed in the

Table 4 Pearson's correlation coefficients for the trace element contents and sediment properties in the study area

	Fe	Pb	Cd	Zn	Ni	Co	Cr	Cu	Mn	Sand	Silt	Clay	pH	TOM	CEC
Fe	1	0.26	0.37	0.4	0.3	0.22	0.43	0.25	0.22	−0.39	0.18	0.32	0.32	0.22	0.24
Pb		1	**0.79**	**0.84**	**0.65**	**0.83**	**0.84**	**0.86**	**0.73**	**−0.83**	0.34	**0.73**	**0.51**	**0.89**	**0.74**
Cd			1	**0.84**	**0.68**	**0.73**	**0.78**	**0.79**	**0.67**	**−0.79**	0.15	**0.86**	0.44	**0.83**	**0.81**
Zn				1	**0.74**	**0.78**	**0.92**	**0.89**	**0.7**	**−0.97**	**0.52**	**0.8**	**0.5**	**0.96**	**0.79**
Ni					1	**0.64**	**0.56**	**0.65**	**0.75**	**−0.67**	0.21	**0.69**	0.45	**0.7**	**0.69**
Co						1	**0.78**	**0.88**	**0.8**	**−0.75**	0.18	**0.8**	**0.64**	**0.8**	**0.85**
Cr							1	**0.9**	**0.68**	**−0.94**	**0.52**	**0.74**	0.47	**0.9**	**0.79**
Cu								1	**0.73**	**−0.88**	0.31	**0.85**	**0.53**	**0.9**	**0.86**
Mn									1	**−0.7**	0.25	**0.7**	**0.54**	**0.76**	**0.79**

Significant correlation coefficients (at $p<0.01$) are in bold

dendrogram are between Zn and Cr. These results are supported by Pearson's correlation coefficients (Table 4). Strongly positive correlations were observed among the elements in cluster C1; i.e., between Zn and Cr ($r=0.92$), Zn and Pb ($r=0.84$), Zn and Cd ($r=0.84$), Cd and Pb ($r=0.79$), Cr and Pb ($r=0.84$), Cr and Cd ($r=0.78$), Cu and Pb ($r=0.86$), Cu and Cd ($r=0.79$), and Cu and Zn ($r=0.89$). In addition, positively strong correlations were found among the components of cluster C2 (i.e., between Co and Mn, Co and Ni, and Ni and Mn, with $r=0.80$, 0.64, and 0.75, respectively) (Table 4).

The results of the PCA are presented in Table 5. Two principal components are extracted, which account for 87.75 % of the total variance. The first factor (PC1) explains 48.4 % of the total variance, and contains a high positive loading of Pb, Co, Cu, and Mn. The second factor (PC2), accounting for 39.3 % of the total variance, shows highly positive loading for Cd and Zn.

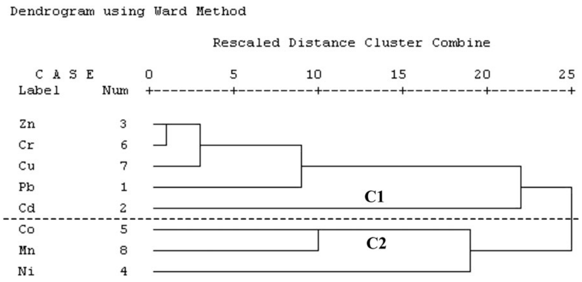

Fig. 4 The dendrogram of hierarchical cluster analysis (HCA) for the studied trace metal concentrations in surface sediments of Ziqlab reservoir

Assessment of sediment contamination

Assessment of trace element pollution using sediment quality guidelines

The extent of trace element pollution in surface sediments of Ziqlab dam was assessed by the sediment quality guidelines (SQG) (Giesy and Hoke 1990). Based on the SQG guidelines, all bottom sediments fall in the non-polluted and moderately polluted classes, except for Ni and Mn (Table 3), where Ni also showed higher

Table 5 Principal component loadings and explained variance for the two components with Varimax normalized rotation

Element	PC1	PC2
Pb	**0.76**	0.54
Cd	0.30	**0.91**
Zn	0.63	**0.74**
Ni	0.60	0.60
Co	**0.84**	0.40
Cr	0.67	0.66
Cu	**0.70**	0.65
Mn	**0.89**	0.32
Percent of variance	48.40	39.35
Cumulative percent	48.40	87.75

Bold values=loadings>0.7

values indicating heavily polluted sediments, while the low Mn contents indicated only non-contaminated sediments. Ninety, 87, 53, and 37 % of sediments were non-polluted with Pb, Cu, Zn, and Cr, respectively, whereas the remaining samples were moderately polluted with these metals. However, all samples were found with Mn concentrations of less than 300 mg kg^{-1}, below which sediment is considered non-polluted. Ni is the only metal with higher values indicating heavily polluted sediments (27 %). The majority of sediments were moderately contaminated with Ni (60 %), whereas 13 % of surface sediments of Ziqlab dam were non-polluted with Ni.

It is noteworthy to mention that the SQGs (developed by the US-EPA) do not consider natural background or multiple metals, which is especially important for trace elements that may occur naturally at greater concentrations in some areas of the world.

Assessment of trace element pollution using geoaccumulation index

The geoaccumulation index (Igeo) is a common criterion used for evaluating the trace metal pollution in sediments (Leopold et al. 2008). It was originally defined by Muller (1979), where trace metal contamination in sediments was determined by comparing their current concentration levels with those from preindustrial times. The Igeo can be defined as:

$$\mathrm{Igeo} = \log_2 \left(Cn \middle/ (1.5 \times Bn) \right) \quad (1)$$

where Cn is the measured concentration of metal (n) in the sediments, Bn is the geochemical background concentration of the metal (n) in shale (Turekian and Wedepohl 1961), and 1.5 is a background matrix correction factor for lithogenic effects. Muller (1981) determined seven classes of Igeo in sediments: (i) Igeo<0 for unpolluted, (ii) 0<Igeo<1 for unpolluted to moderately polluted, (iii) 1<Igeo<2 for moderately polluted, (iv) 2<Igeo<3 for moderately to strongly polluted, (v) 3<Igeo>4 for strongly polluted, (vi) 4<Igeo<5 for strongly to very strongly polluted, and (vii) Igeo>5 for very strongly polluted conditions.

The basic statistics of Igeo calculations for trace metals are listed in Table 6 and their spatial variability in the reservoir sediments are presented in Fig. 5. Based on Muller scales (Muller 1981), reservoir's sediments are unpolluted to moderately polluted with Cd (i.e., Igeo ranging from 0.000 to 2.080) (Table 6). Pb levels in sediments at 13 sites (43.3 %) were classified as unpolluted to moderately polluted (0<Igeo<1), but 56.7 % of total sites (17 sites) were unpolluted (Igeo<0). All other metals (Cu, Zn, Ni, Cr, Co, and Mn) in the studied samples showed lower values (Igeo<0) indicating that the reservoir surface sediments are unpolluted. On the basis of mean Igeo values, trace metals were ranked in the following order: Cd (0.704)>Pb (−0.500)>Zn (−1.223)>Ni (−1.360)>Co (−1.643)>Cr (−2.082)>Cu (−2.202)>Mn (−4.682).

Figure 5 shows the spatial trends of Igeo for Pb, Cd, Cu, Zn, Ni, Co, Cr, and Mn in surface sediments of the Ziqlab reservoir. The majority of trace metals showed similar distribution patterns; i.e., more positive Igeo values in the western side (lake outlet), compared to the eastern reservoir (next to lake inlet). This may be due to the high loading of these metal concentrations in the western side, where clayey-silt facies dominate (Fig. 1). The Cd and Pb values showed a remarkably positive behavior with few variations in Igeo across the sampling sites.

Assessment of trace element pollution using enrichment factor

Enrichment factor (EF) analysis is a method proposed by Simex and Helz (1981) to assess trace element concentrations. It is mathematically expressed as:

$$\mathrm{EF} = \frac{\left(\mathrm{M}/\mathrm{Fe}\right)_{\text{sample}}}{\left(\mathrm{M}/\mathrm{Fe}\right)_{\text{crust}}} \quad (2)$$

where $(\mathrm{M/Fe})_{\text{sample}}$ is the ratio of metal and Fe concentrations in the sample, and $(\mathrm{M/Fe})_{\text{crust}}$ is the ratio of metal and Fe concentrations in the Earth's crust.

The EF values for the trace metals are tabulated in Table 6. According to Zhang and Liu (2002), elements with EF<2 are considered to entirely originate from the crustal materials or natural processes, while those elements with EF>2 are most likely the product of anthropogenic activities. The high EF values for Cd and Pb (with an average of 5.83 for Cd and 3.05 for Pb) (Table 6), indicating anthropogenic impact on metal concentration in

Table 6 Geoaccumulation index (Igeo) and enrichment factor (EF) of trace metals in sediments

	Pb	Cd	Cu	Zn	Ni	Cr	Co	Mn
Igeo								
Minimum	−3.91	0	−4.08	−3.83	−2.97	−5.49	−3.83	−6.86
Maximum	0.71	2.08	−1.12	−0.25	−0.21	−1.05	−0.66	−3.67
S.D.	1.13	0.71	0.796	0.999	0.8	1.08	0.67	0.944
Average	−0.5	0.7	−2.2	−1.22	−1.36	−2.08	−1.64	−4.68
EF								
Minimum	0.31	0.82	0.02	0.22	0.45	0.07	0.26	0.03
Maximum	5.23	13.5	0.15	3.46	3.2	1.68	2.02	0.3
S.D.	1.41	3.74	0.036	0.85	0.77	0.432	0.432	0.075
Average	3.05	5.83	0.087	1.795	1.75	0.993	1.24	0.162

SD standard deviation

reservoir sediments. The remaining metals showed EF values of less than 2 (i.e., Zn (1.795)>Ni (1.753)>Co (1.240)>Cr (0.993)>Mn (0.160)>Cu (0.087)). These values suggest that they were originated from natural sources.

The EF for all metals in surface sediments of Ziqlab reservoir showed similar variations, with more positive EF values found in the western reservoir (outlet), compared to the eastern portion of reservoir (inlet) (Fig. 6). This pattern of EF values is associated with dominant clayey-silt facies in the western part (Fig. 1), along with high percent of TOM (Fig. 2).

Pollution sources

The results of the PCA analysis for total trace metal concentrations in surface sediments are listed in Table 5. The factors explain a relatively large extent of the total variance (87.75 %) of the eight variables used in this analysis. The first factor (PC1), which explains 48.40 % of the total variance, shows highly positive loading for Pb, Co, Cu, and Mn (Table 5). This indicates that the concentrations of these metals are closely related and that they were originated from similar sources (probably of lithologic origin) or comparable chemical properties (Hakanson and Jasson 1983). The correlation analysis also shows highly significant positive correlations ($P<0.01$) among the first factor (PC1) elements (Table 4). However, effluent and leakage of landfill and animal wastes from the surrounding area may have probably contributed to the high metal concentrations, particularly for Pb (El-Radaideh et al. 2014) which is consistent with the high Igeo and EF values observed for Pb (Table 6).

The second factor (PC2), which accounts for 39.35 % of the total variance, shows highly positive factor loading for Cd and Zn (Table 5). The correlation analysis indicates a highly significant positive correlation between Cd and Zn (0.84) (Table 4). Cd and Zn concentrations may be elevated because of inputs from agricultural sources, including fertilizer use in the upstream agricultural farms along the main wadies (Abu-Rukah and Ghrefat 2004; El-Radaideh et al. 2014). Abed et al. (2008) argued that Cd and other potentially toxic trace metals are enriched in the diammonium phosphate (DAP) fertilizer by a factor of about 2 and a factor of 3 in the phosphoric acid. However, Abed (1982) and Jarrar and Mustafa (1995) showed that the weathering of the lithological units exposed at the dam site is the major source of trace metals, particularly limestone, bituminous limestone, basalt, and chalky marl. High Zn levels (with an average of 0.274 mg/g dry wt.) were reported by Abed (1982) in the bituminous limestone. Jarrar and Mustafa (1995) also showed Zn and Cd contents in the bituminous limestone that varied from 0.135 to 0.532 mg/g dry wt. (with an average of 0.324 mg/g dry wt.), and from 0.007 to 0.034 mg/g dry wt. (with an average of 0.016 mg/g dry wt.), respectively.

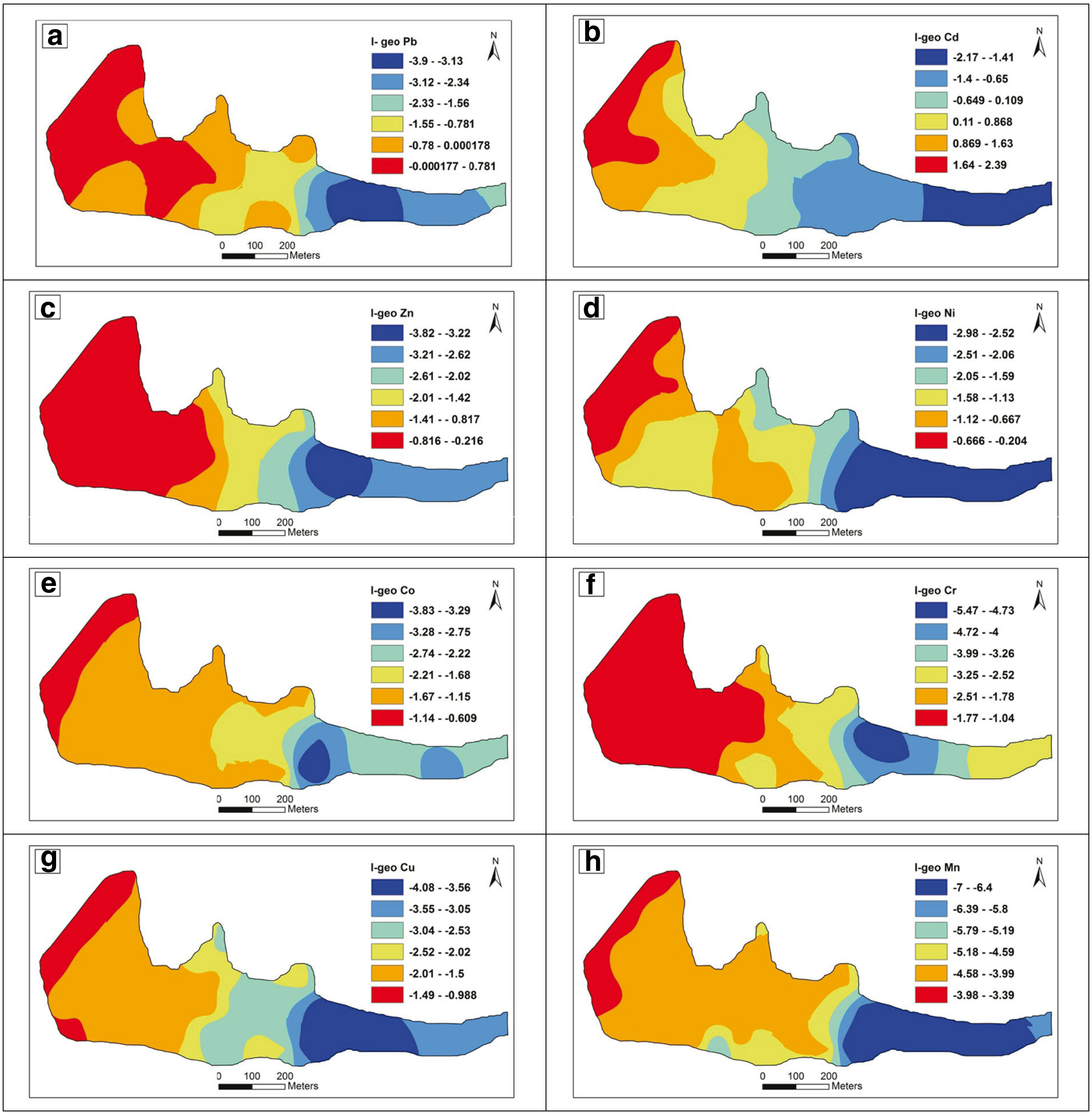

Fig. 5 Spatial distribution of geoaccumulation index (Igeo) for Pb (**a**), Cd (**b**), Cu (**c**), Zn (**d**), Ni (**e**), Co (**f**), Cr (**g**), and Mn (**h**) in surface sediments of Ziqlab reservoir

Environmental factors that influence the mobility of metals, such as pH, TOM, and CEC may also explain the variations in trace element levels (Laing et al. 2008). Soil pH is positively correlated with Pb, Zn, Co, Cu, and Mn (Table 4), which is associated with the low mobility of these elements in the alkaline environment (Gao et al. 2013). Zeng et al. (2001) also reported a negative correlation between soil pH and extractable trace element levels. Thus, higher soil pH values would lead to the accumulation of these elements in soil. Table 4 shows significant positive correlations between TOM and CEC and all of the studied trace elements.

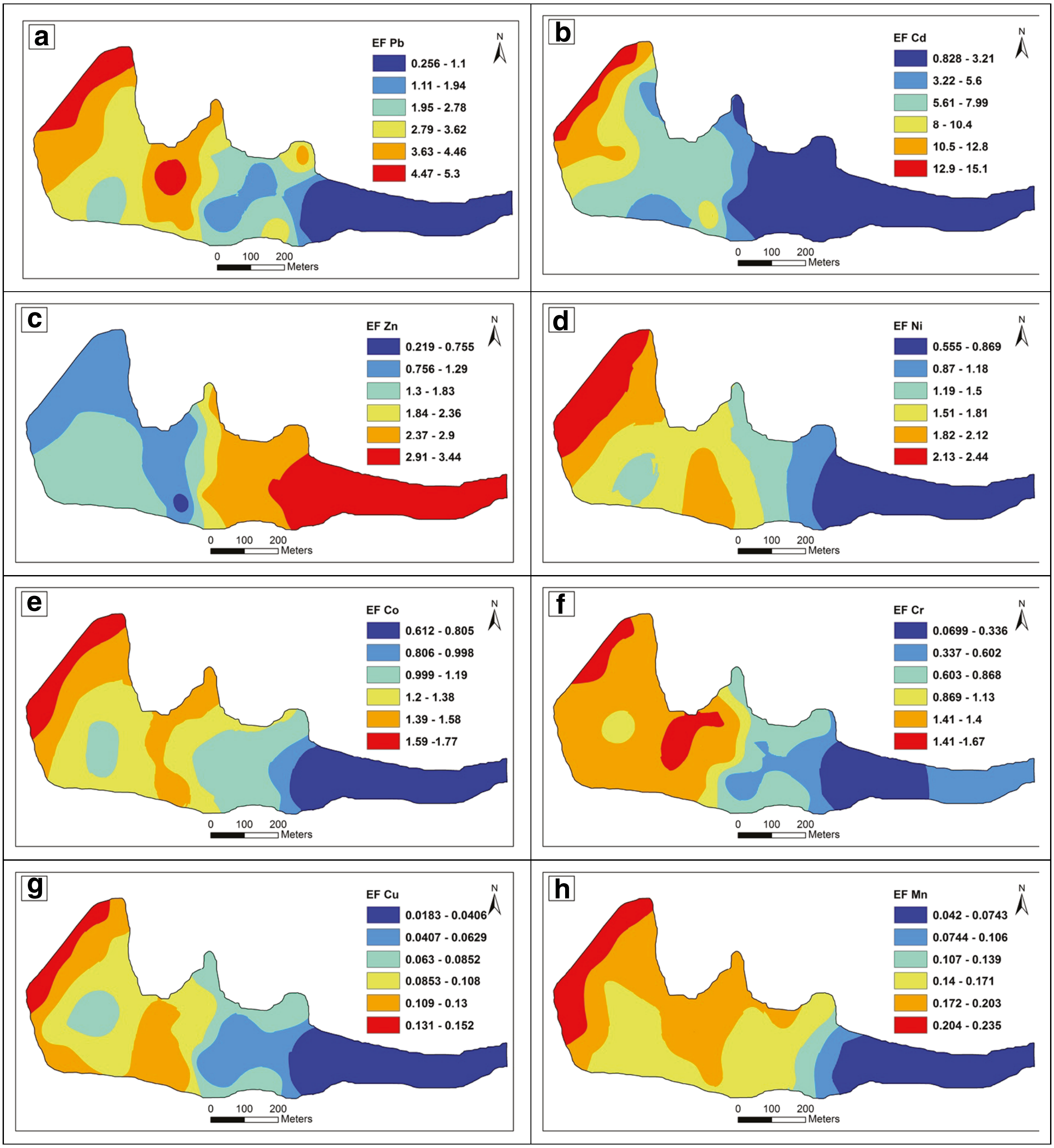

Fig. 6 Spatial distribution patterns of enrichment factor (EF) for Pb (**a**), Cd (**b**), Cu (**c**), Zn (**d**), Ni (**e**), Co (**f**), Cr (**g**), and Mn (**h**) in surface sediments of Ziqlab reservoir

Conclusions

This study focuses on the spatial distribution and contamination levels of trace metals in surface sediments from Ziqlab dam in northwestern Jordan. Trace metal concentrations varied spatially with relatively similar distribution patterns. These variations were closely linked to the TOM and fine fraction distributions. Weathering of rocks in the catchment and agricultural activities are the primary sources of trace metal in the surface sediments. The levels of Pb, Cd, and Zn exceeded the environmental background guidelines.

Based on the US-EPA SQGs, surface sediments are considered non-polluted to moderately polluted with the majority of metals, except for Ni which also showed high values indicating heavily polluted sediment. The Igeo of trace metals are in the following order: Cd > Pb > Zn > Ni > Co > Cr > Cu > Mn. The bottom sediments were enriched in Cd and Pb indicating their anthropogenic origins, whereas low enrichment values of the remaining elements suggesting their natural sources.

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