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Seasonal variation of heavy metal concentrations in sediments of Farwa Lagoon, Libya

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A B S T R A C T

This study aimed to investigate the seasonal variations in the concentrations of some heavy metals (cadmium, lead, copper, manganese, nickel, and zinc) in the surface sediments of Fatwa lagoon. Six different stations were selected as sampling points. Samples were taken over three seasons, and seasonal and annual average element concentrations were determined. It was observed that the seasonal concentrations of heavy metals (lead, copper, manganese, nickel, and zinc) during summer, autumn, and winter were within the normal range in the surface sediments and that the lagoon was not contaminated with these metals. However, cadmium concentrations in the surface lagoon sediments were found to exceed the permissible limits during the rainy season.

Keywords: *Farwa lagoon, seasonal variations, permissible limits, Heavy metals, sediments*

التباين الموسمي لتركيزات المعادن الثقيلة في رواسب بحيرة فروة

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الملخص

هدفت هذه الدراسة إلى التعرف على التغيرات الموسمية في تركيزات بعض المعادن الثقيلة (الكادميوم، والرصاص، والنحاس، والمنجنيز، والنيكل، والزنك) في الرواسب السطحية لبحيرة فروه. اختيرت ست محطات مختلفة كنقاط لأخذ العينات. أُخذت العينات على مدار ثلاثة مواسم، وحُدد متوسط تركيزات العناصر الموسمية والسنوية. لوحظ أن التركيزات الموسمية للمعادن الثقيلة (الرصاص، والنحاس، والمنجنيز، والنيكل، والزنك) خلال فصول الصيف والخريف والشتاء كانت ضمن المعدل الطبيعي في الرواسب، وأن البحيرة لم تكن ملوثة بهذه المعادن. ومع ذلك، وُجد أن تركيزات الكادميوم فى رواسب البحيرة السطحية تتجاوز الحدود المسموح بها خلال الموسم الممطر.

الكلمات المفتاحية: *بحيرة فروه، التباين الموسمى، الحدود المسموح بها، المعادن الثقيلة، الرواسب.*

1. Introduction

Coastal lagoons function as transitional environments that connect continental and marine ecosystems, receiving biogeochemically active contributions from land, rivers, and marine coasts; they are among the most productive ecosystems in the biosphere and are characterized by their great complexity due to their position connecting land and sea [1]. These ecosystems are controlled by complex interactions among land, ocean, and atmosphere, leading to highly variable and vulnerable environments [2]. Numerous types of pollutants exist in the environment, including organic materials, major ions, and trace metals, which can enter the aquatic ecosystem due to urbanization, industrial activities, and agriculture [3]. Heavy metals are toxic and pose significant threats to the environment due to their long persistence and resistance to oxidation, degradation, removal, or conversion into less harmful components through biological or chemical processes. Although there is no strict definition of a limit, heavy metals are typically characterized by their density, atomic weight, atomic number, or position in the periodic table [4]. Various human activities, such as smelting, mining, electroplating, and other industrial operations, release metals into their wastewater, potentially disrupting the ecological balance of the region. Some elements, such as Mn, Fe, Zn, and Cu, are essential for aquatic organisms. Others, including Hg, As, and Pb, can be toxic to these organisms, even at trace concentrations. As a result, the types and quantities of metals entering the environment must be considered when analyzing potential harmful impacts [5]. Monitoring metal levels offers valuable indicators of anthropogenic inputs to assess metal pollution and predict its impact on ecosystems, as heavy metals can easily bioaccumulate in organisms. Sediments are sensitive

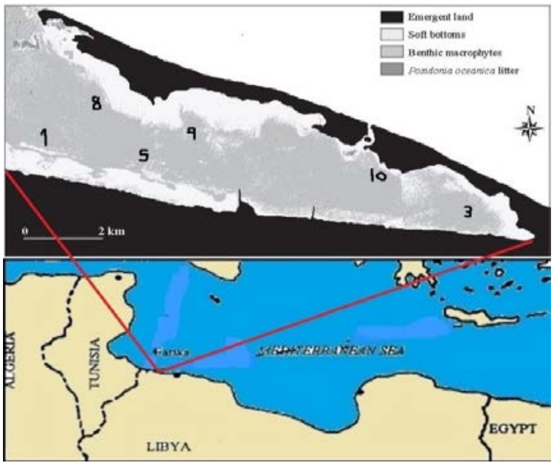

Figure 1 Sampling sites in Farwa lagoon Sampling and analysis

indicators for monitoring contaminants in aquatic ecosystems and are often regarded as the ultimate sink for heavy metals deposited into these environments [6]. Understanding the concentrations and distribution of trace metals in sediments can thus be crucial in identifying sources of contaminants in aquatic systems. Numerous studies across various regions of the world have utilized the sediments of rivers, estuaries, and seas as indicators of trace metal pollution [7]. The purpose of this study was to determine the levels of heavy metals in Farwa Lagoon sediments to assess pollution levels in the area. This study also seeks to contribute to the creation of a database of these metals in this area.

2. Material and methods

Study area

The study area, Farwa Lagoon, is located 150 km west of Tripoli and has an area of about 310 hectares. The lake is separated from the sea by a narrow strip 11 km long. The lagoon communicates with the sea through two openings, one of which is located west of the lagoon and is relatively wide, 3.5 km wide, and the other is located east of the lagoon and is 10 meters wide. The depth of the lagoon ranges from 0.5 to 2.5 meters. Figure 1 shows the studied area (Farwa lagoon) and the sample collection stations. Sediment samples were collected from 6 stations, three times, to determine environmental changes during the dry season and the rainy season (July 2010, October 2010, and January 2011). The upper 5 cm of sediment was collected and placed in a plastic container that had been previously cleaned and rinsed with distilled water. The plastic containers were frozen in the field and transported to the laboratory for analysis. The sediments were dried at 60°C for 24 h. Samples were sieved through a 200 µm to 63 µm mesh. A portion of each sediment sample with a size of > 63 µm mesh was dried at 105°C for 24 h to constant weight and homogenized before analysis. Acid-washed glassware, Analytical-grade reagents and double-distilled deionized water were used throughout the experiments. Standard stock solutions of Cu, Cd, Pb, Ni, Mn, and Zn were prepared from Fluka (1000 mg/l). The working solutions were freshly prepared by diluting an appropriate aliquot of the stock solutions. Sediment samples were digested using a Start D Microwave. Table 1, represents the microwave program for sea sediments. Application field: Environment.

Sample amount

0.25 g

Reagents

6 ml of HNO_3 65%, 1 ml of H_2O_2 30% and 1 ml of $HClO_4$ 65%

Table 1. Microwave program

Step	Time(min)	Temperature	Microwave power
1	10	200°C	Up to 1000 Watt
2	20	200°C	Up to 1000 Watt

Metals were analyzed using Atomic Absorption Spectrometry (M SERIES) from Thermo Fisher Scientific. One-way ANOVA was used to evaluate

Table 2. Concentration of metals in mg/l (dry weight) in the sediments of Farwa Lagoon in July 2010

station	Cu	Cd	Pb	Ni	Mn	Zn
1	1.712	0.105	1.473	1.131	39.62	18.66
3	2.461	0.177	0.470	0.962	45.67	16.45
5	3.214	0.121	1.201	0.620	32.42	17.32
8	2.671	0.066	1.860	1.271	62.68	22.13
9	0.979	0.163	1.091	0.673	40.31	22.06
10	3.888	0.108	1.618	0.973	29.56	18.1
AVE	2.488	0.123	1.286	0.938	41.71	19.12
STDEV	1.039	0.041	0.487	0.253	11.79	2.424

Table 3. Concentration of metals in mg/l (dry weight) in the sediments of Farwa Lagoon in October 2010

station	Cu	Cd	Pb	Ni	Mn	Zn
1	2.292	4.926	4.892	0.577	20.67	16.06
3	2.830	12.68	3.227	0.931	39.91	17.39
5	3.872	10.83	4.509	0.317	29.67	16.92
8	3.453	8.715	8.132	1.514	22.83	28.69
9	1.857	12.205	1.943	0.175	35.09	27.10
10	5.056	5.088	5.341	0.881	39.04	19.77
AVE	3.226	9.075	4.674	0.733	31.20	20.98
STDEV	1.159	3.440	2.099	0.486	8.194	5.515

Table 4. Concentration of metals in mg/l (dry weight) in the sediments of Farwa Lagoon in January 2011

station	Cu	Cd	Pb	Ni	Mn	Zn
1	2.475	5.214	4.011	0.511	38.63	18.12
3	2.824	11.08	3.984	0.887	40.02	19.08
5	3.711	10.88	4.734	0.309	33.33	17.21
8	4.017	8.114	7.321	1.354	45.28	29.43
9	2.146	11.43	2.320	0.758	42.31	23.94
10	4.726	4.782	5.081	0.871	39.23	21.91
AVE	3.316	8.585	4.575	0.782	39.80	21.62
STDEV	0.995	3.023	1.648	0.359	3.997	4.575

the statistical differences between the means of the metals from the study sites using SPSS software version 25.

3. Results and discussion

A. Results

Heavy metals from both natural and anthropogenic sources accumulate in sediments and plants, consequently posing significant environmental contamination issues [8].

Copper (Cu): From the results obtained in Tables 2, 3, and 4, it was observed that the copper concentration during the collection seasons ranged from 0.979 to 5.056 with an annual average of 3.010 ± 0.085 mg/l. The highest concentration of 5.056 mg/l was recorded at Station (10) during the rainy season in October 2010, while the lowest concentration of 0.979 mg/l was recorded at Station (9) during the dry season in July 2011. In general, the copper concentration values for all stations during the collection seasons were found to be close. The results of the analysis of variance (ANOVA) concerning seasonal variations in copper show no significant seasonal variation ($p > 0.05$, $p = 0.361$).

Cadmium (Cd): The primary sources of cadmium pollution are the metal and plastics industries, as well as sewage. A significant portion of cadmium (30-50%) is located in the most mobile forms (either exchangeable or carbonate-bound), allowing it to easily enter the food chain [9]. According to the results presented in Tables 2, 3, and 4, the cadmium concentration during the collection seasons varied from 0.066 to 12.687 mg/l, with an annual average of 5.928 ± 1.854 mg/l. The highest concentration was recorded during the rainy season at station (3), where the water was shallow and low. The lowest cadmium concentration was noted at station (8) in the dry season. It was found that cadmium concentrations in the rainy seasons exceeded those in the dry season across all collection stations. The results of the analysis of variance (ANOVA) regarding seasonal variations in cadmium indicate highly significant seasonal variation ($p<0.05$, $p=0.000$).

Lead (Pb): A significant portion of Pb in sediment is linked to exchangeable and residual fractions, allowing it to easily enter the food chain [10]. From the results shown in Tables 2, 3, and 4 it is clear that the concentration of lead in the sediments of Farwa

Table 5. Sediment quality guideline (SQG) by US EPA [11, 12, 13]

Metals	Not polluted	Moderately polluted	Heavily polluted	Present study
Cd	-	-	> 6	0.066-12.68
Cu	<25	25 – 50	> 50	0.979-5.05
Pb	<40	40 – 60	> 60	0.47-7.32
Ni	<20	20 – 50	> 50	0.175-1.51
Mn	<300	300 – 500	> 500	20.67-62.67
Zn	<90	90 - 200	> 200	16.44-29.43

Table 6. [14, 15, 16, 17].

Permissible limits worldwide	Cd	Cu	Pb	Ni	Mn	Zn
EU (2002)	3	300	300	75	-	300
CSQGD (2007)	1.4	70	70	50	-	-
US EPA (1999)	0.01-4.1	19	19	19	550	60

Lagoon ranged from 0.47 to 8.132 mg/l, with an annual average of 1.928 ± 0.832 mg/l, where Station (8) recorded the highest concentration during the rainy season, and Station (3) showed the lowest concentration during the dry season. All sampling stations showed higher lead values during the rainy season compared to the values recorded during the dry season. The results of the analysis of variance (ANOVA) regarding seasonal variations in lead indicate very significant seasonal variation ($p<0.05$, $p=0.003$).

Nickel (Ni): The results indicate that the nickel values obtained from all stations during the sampling seasons were similar, with no significant differences observed among them. Nickel concentrations ranged from 0.620 to 1.514 mg/l during the sampling seasons, with an annual average of 0.818±0.116 mg/l. The highest concentration of nickel was recorded at station (8) during the rainy season, and the lowest concentration was found at station (5) during the dry season. The results of the analysis of variance (ANOVA) regarding seasonal variations in lead indicate no significant seasonal variation ($p>0.05$, $p=0.625$).

Manganese (Mn): The manganese concentration ranged during the sampling seasons from 20.67 to 62.68 mg/l as shown in Tables 2, 3, and 4 with an annual average of 37.57 ± 3.601 mg/l. The highest manganese value was recorded at station (8) during the dry season, while the lowest was noted at station (1) in autumn. The results of the analysis of variance (ANOVA) regarding seasonal variations in manganese indicate no significant seasonal variation ($p>0.05$, $p=0.113$).

Zinc (Zn): The results from Tables 2, 3, and 4 indicated that there was no significant difference in zinc concentrations across the sampling seasons. The zinc concentration in the sediments of Farwa Lagoon ranges during the collection seasons from 16.055 to 29.43 mg/l with an annual average of 20.58 ± 1.585 mg/l. The highest concentration was observed at station (8) during the rainy season. The analysis of variance (ANOVA) results concerning

Table 7 Heavy metal levels in sediment samples from the Libyan Mediterranean coast and other selected regions of the Mediterranean Sea.

Region	Cu	Cd	Pb	Ni	Mn	Zn	Ref.
Libya	0.98-5.1	0.18-12	0.47-8.1	0.62-1.5	29.6- 45	16.5-29	Present study
Libya	3.4-4.3	0.3-0.8	0.16-1.1	-	32.6-94	4.0-7.9	24
Libya	9.1-22.7	5-10.5	18.9-57	11.6-30	14.3-49	11.6-30	25
Tunis	27.3	1.51	102	71.8	-	148	26
Italy	4.4-21.7	0.2-0.94	5.2-7.7	0.2-15	-	48.3-95	27
France	19.7	0.14	28	-	365	72	28
Greece	0.5-198	0.16-1.6	20-282	26-296	1.1-905	2.3-599	29
Egypt	16.9	3.88	50.6	45.6	-	67.2	30

seasonal variations in manganese show no significant seasonal variation (p>0.05, p=0.599).

B. Discussion

The annual average of the studied ecosystem was as follows: Cd: 5.928 µg/g; Cu: 3.010 µg/g; Pb: 3.51 µg/g; Mn: 37.57 µg/g; Ni: 0.818 µg/g; and Zn: 20.57 µg/g dry weight. This allows for the arrangement of the metals from higher to lower mean content in this area as: Mn > Zn > Cd > Pb > Cu > Ni.

Examining Tables 5 and 6, all heavy metals measured in the sediments were within permissible limits, except for cadmium, which showed an increase at sampling stations (3-5-8-9) during the rainy season. In a similar study conducted by [22], it was shown that the studied sites are severely contaminated with cadmium, causing very high environmental risks that affect the quality of the sediments of Farwa Lagoon. In a study conducted by [23], the results clearly showed that the average concentration of cadmium and lead was higher than the permissible level in samples near the wastewater treatment plant, indicating that this station may be the most important reason for the increase in metal concentrations in the study area. According to [18], the highest concentrations of cadmium and nickel were recorded during the rainy season. This can be attributed to the influx of these elements into the lake through surface runoff. This increase in cadmium may be due to the contribution of industrial and domestic wastewater from urban areas and rainwater runoff [19]. The presence of Cd in the environment warrants attention due to its high toxicity and its accumulation in sediments, which is entirely attributed to anthropogenic activities involving the use of this metal [20].

A comparison of heavy metal concentrations in sediments from various Mediterranean regions with the findings of this study Table 7 indicates that Farwa Lagoon exhibited the highest cadmium concentration, which aligns with the findings reported in [22]. Additionally, the results of this study are consistent with [25] regarding the elevated levels of cadmium concentration. The increase in heavy metal concentrations occurs due to the interference of industrial and domestic wastewater [26]. The high metal content of Lake Farwa's sediments may be due to the blockage of the channel to the northeast of the lake by seaweed and sediment accumulation. This prevents the lagoon from properly replenishing its water supply, increasing water concentration and causing it to rot, creating a polluted environment.

4. Correlation Coefficient Analysis of Heavy Metals.

High levels of association between heavy metals in sediments may indicate similar contamination

Table 8. Correlation Coefficient Analysis of Heavy Metals during summer.

	Cu	Cd	Pb	Ni	Mn	Zn
Cu	1.0					
Cd	-0.4	1.0				
Pb	0.3	-0.9	1.0			
Ni	0.1	-0.6	0.5	1.0		
Mn	-0.3	-0.3	0.2	0.6	1.0	
Zn	-0.5	-0.3	0.5	0.2	0.6	1.0

Table 9. Correlation Coefficient Analysis of Heavy Metals during winter.

	Cu	Cd	Pb	Ni	Mn	Zn
Cu	1.0					
Cd	0.6	1.0				
Pb	0.7*	-0.4	1.0			
Ni	0.3	-0.1	0.6	1.0		
Mn	-0.1	-0.1	0.3	0.9*	1.0	
Zn	0.2	-0.04	0.5	0.9*	0.9*	1.0

and/or pollution originating from the same sources [17].

The correlation coefficient matrix for the selected heavy metals is shown in Tables 8 and 9. During the summer, there are no significant correlations among most of these heavy metals, indicating that they are not associated with one another and may have different anthropogenic and natural sources in the sediments of the study area. In winter, significant correlations between the metals Ni and Mn (r=0.9), Cu and Pb (r=0.7), Ni and Zn (r=0.9), and Mn and Zn (r=0.9) may suggest a common or similar source of input [13].

5. Conclusion

Understanding heavy metal concentrations in marine sediment is crucial for natural management, aquatic ecology, and human health [21]. This study showed that the lake sediments were free of the studied heavy metals, except for an increase in cadmium concentrations during the rainy season. This increase may result from soil erosion within the lake, in addition to the possibility of sewage seeping into the lake. This research contributes to the creation of a database of heavy metal concentrations, which will serve as a reference for future studies in the region.

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