

Assessment of Metal(loid) Accumulation in the Surficial Sediment of Meyil Lake

Meyil Gölü Yüzeysel Sedimentinde Metal(loid) Birikiminin Değerlendirilmesi

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ABSTRACT

This study investigated eight metal (Cu, Pb, Zn, Ni, Mn, Fe, Cr, Al) and one metalloid (As) amounts in the sediment of Meyil Sinkhole Lake. The findings obtained were compared with the limit values of sediment quality guidelines (SQGs) such as PEL (probable effect level), TEL (threshold effect level), ERL (effects range low) and ERM (effects range median). In addition its probable toxic effects were investigated via different sediment evaluation methods. As a result, the value of the metal concentrations tested was below the limit value. The contamination factor of all the metal(loid)s tested was found to be below 1 ($C_f^i < 1$). In other words, the accumulation level of all metal(loid) was found to be low. Also the mean ERM quotient (m-ERM-q) and mean PEL quotient (m-PEL-q) values revealed that there was no risk for the lake. The total toxic unit (Σ TU) method revealing the toxic effect of the metal(loid)s, also supported this result. In the light of all these data, the sediment of the lake was found to be clean in terms of investigated metal(loid). The accumulation relations of metals (loids) are also revealed by correlation analysis. The correlations between Zn-Pb and Fe were remarkable

Keywords: Sinkhole, karst lake, sediment quality guidelines

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ÖZET

Bu çalışmada Meyil Obruk Gölü sedimentinde 8 metal (Cu, Pb, Zn, Ni, Mn, Fe, Cr, Al) ve 1 metalloid'in (As), miktarları araştırılmıştır. Elde edilen bulgular PEL (muhtemel etki seviyesi), TEL (eşik etki seviyesi), ERL (düşük etki aralığı), ERM (ortalama etki aralığı) gibi sediment kalite rehberi (SQGs) sınır değerleri ile karşılaştırılmıştır. Ayrıca farklı sediment değerlendirme yöntemleri ile de muhtemel toksik etkileri araştırılmıştır. Elde edilen sonuçlar neticesinde test edilen metal konsantrasyonlarının değeri sınır değerlerin altında olduğu tespit edilmiştir. Test edilen tüm metal(loid)lerin kontaminasyon faktörü 1'in ($C_f^i < 1$) altında bulunmuştur. Yani tüm metal(loid)lerin birikim seviyesi oldukça düşük bulunmuştur. Ayrıca ortalama etki aralığı medyanı oranı (m-ERM-Q) ve ortalama muhtemel etki seviyesi oranı (m-PEL-Q) değerleri göl için herhangi bir riskin olmadığını ortaya koymuştur. Bu sonuç metal(loid)lerin toksik etkisini ortaya koyan toplam toksik ünite (ΣTU) yöntemi ile de desteklenmiştir. Tüm bu veriler ışığında gölün sedimenti araştırılmış meta(loid)ler açısından temiz bulunmuştur. Metal(loid)lerin birikim ilişkileri de korelasyon analizi ile ortaya konmuştur. Zn-Pb ve Fe arasındaki korelasyonlar dikkat çekici bulunmuştur.

Anahtar Kelimeler: Obruk, karstik göl, sediment kalite rehberi

1. INTRODUCTION

Sinkholes are carstic wells which vertically go down deep and look like a large chimney (Erinç, 2012). They have complicated environmental features due to their unique geomorphology (Hofierka *et al.*, 2018). The Meyil Lake is one of the sinkhole lakes positioned in the Karapınar Plain in the direction of Eskişehir district (37°59'17.83"N -33°21'12.98"E). The diameter of the lake which is 60 meters deep from the ground level, ranges from 200 to 300 meters. In the water of the lake which is 40 meters deep, there are wide varieties of fish and zooplankton (Tapur and Bozyiğit, 2016; Durmaz, 2019).

There is a direct correlation between rainfalls and underground waters and between underground water level and sinkhole formations. The number of wells drilled to meet the water need of the increasing irrigated farming in Konya Karapınar area, is gradually increasing. Unfortunately these irrigation wells which are drilled unconsciously, cause a decrease in the underground water level and water pollution (such as pesticides, heavy metals) (Yılmaz, 2010). Sinkholes are open systems to the circulation of heavy metals between underground waters and surface waters due to the cracks (dolines, channels and caves) in their structure (Lang *et al.*, 2006; Wu *et al.*, 2014).

Heavy metals are among pollutants that are

accepted to be a source of concern worldwide due to their environmental permanence and ecologic risks. The main pool of heavy metals in wetlands is sediment and pollution in sediments is an important indicator of water pollution (Nguyen *et al.*, 2019).

It has been reported that metals (such as chrome, nickel, cadmium, mercury and lead) tend to precipitate as hydroxides and carbonates in the water of carbonate-rich carstic areas and the metals here are carried clinging to the surface of small particles such as clays and colloids (Vesper *et al.*, 2003). Thus, assigning the amount of heavy metals in the sediment will provide important information about the area. This information is crucial both for the ecosystem and community health.

There is metal circulation between water and sediment in lakes and this circulation is relatively more intense in shallow lakes (Mrozinska and Bakowska, 2020). Meyil Sinkhole Lake is a relatively shallow and it is also in direct contact with groundwater, which also uses domestic drinking and agricultural irrigation. Therefore, it is very important to investigate the sediment quality of the Meyil Sinkhole Lake for the most common metals.

It is possible to perform the risk assessment of metals detected in a sediment via the Sediment Quality Guidelines (SQG) which contains a series of mathematical formulations (Birch,

2018). The SQG method enables predicting the damage to be caused by anthropogenic effects and interpreting a metal(loid) enrichment, if available (Kwok *et al.*, 2014; Ahamad *et al.*, 2020).

Main goal of the study conducted is to determine the present metal(loid) condition of a sinkhole lake sediment. Also the study aims to determine whether the present condition poses a risk to organisms or not and if it does, which metal(loid)s play a role in this threat and to what extent. Additionally the study aims to determine whether a probable enrichment in metals arises from anthropogenic or natural causes.

2. MATERIALS AND METHODS

Ten surface sediments were collected using plastic materials and were transferred to the laboratory in polyethylene containers (2018) (1-2 cm) (Figure 1). In the laboratory they were kept in a refrigerator under storage conditions of +4 degrees until the analysis. Samples were digested and analyses were determined via the ICP-MS by Çınar Environmental Laboratory. Detection limits of the metal(loid)s were measured as Cu, Pb, Zn, Ni, Mn, Fe, Cr, Al and As; 0.5, 0.5, 5, 0.5, 5, 0.01, 0.5, 0.01 and 5 ppm respectively.



Figure 1. General view of the Meyil Sinkhole Lake (Oğuzhan Durmaz, 2018)

2.1. Assessment Methods for the Anthropogenic Effect

The reference values selected for this study are the source data of Turekian and Wedepohl

(1961). These values are among the most preferred reference values.

2.1.1. Contamination factor (C_f^i)

$$C_f^i = C^i / C_n^i \quad (1)$$

C^i = Quantity of metal

C_n^i = Reference value

wherein:

$C_f^i < 1$ = low contamination factors. $1 \leq C_f^i < 3$ = moderate contamination factors. $3 \leq C_f^i < 6$ = considerable contamination factors and $C_f^i \geq 6$ = high contamination factors (Hakanson, 1980).

2.1.2. Enrichment factor (EF)

$$EF = \frac{C_n / C_{ref}}{B_n / B_{ref}} \quad (2)$$

C_n = Quantity of metal

C_{ref} = Quantity of metal in the reference medium

B_n = Quantity of reference element in the sample

B_{ref} = Quantity of the reference element in the reference medium

Fe is preferred as the reference element.

wherein < 1 = no enrichment. 1 to 3 = minor enrichment. 3 to 5 = moderate enrichment. 5 to 10 = moderately severe enrichment. 10 to 25 = severe enrichment. 25 to 50 = very severe enrichment. > 50 extremely severe enrichment (Hasan *et al.*, 2013).

2.1.3. Geoaccumulation index (I_{geo})

$$I_{geo} = \log_2 \frac{C_n}{1.5 \times B_n} \quad (3)$$

C_n = Quantity of metal

B_n = Quantity of metal in the reference medium

1.5 = natural release coefficient

wherein $I_{geo} \leq 0$ = practically uncontaminated. $0 < I_{geo} < 1$ = uncontaminated to moderately contaminated. $1 < I_{geo} < 2$ = moderately

contaminated. $2 < I_{geo} < 3$ = moderately to strongly contaminated. $3 < I_{geo} < 4$ = strongly contaminated. $4 < I_{geo} < 5$ = strongly to extremely contaminated. $I_{geo} \geq 5$ = extremely contaminated (Müller, 1969).

2.1.4. Ratio of average effects range median (m-ERM-Q) and ratio of average probable effect level (m-PEL-Q)

$$m - ERM - Q = \frac{\sum_{i=1}^n C_i / ERM_i}{n} \quad (4)$$

$$m - PEL - Q = \frac{\sum_{i=1}^n C_i / PEL_i}{n} \quad (5)$$

C_i =Quantity of metal

ERM= effects range median of the metal

PEL= average probable effect level of the metal.

n=number of metal investigated.

wherein $m-ERM-q < 0.1$ = 9%. $0.11 < m-ERM-q < 0.5$ = 21%. $0.51 < m-ERM-q < 1.5$ = 49% and $m-ERM-q > 1.50$ = 76% probability of being toxic (Long *et al.*, 2000).

$m-PEL-Q < 0.1$ = unimpacted. $0.1 < m-PEL-Q < 1$ = moderately impacted. $m-ERM-Q > 1$ = highly impacted (Carr *et al.*, 1996).

2.1.5. Total toxic unit (Σ TU) and relative toxic unit

$$\Sigma TUs = \sum_{i=1}^n C_i / PEL_{C_i} \quad (6)$$

$$Relative TU = \frac{C_i / PEL_{C_i}}{\Sigma TUs} \times 100 \quad (7)$$

Σ TU is the sum of the values obtained by proportioning the quantity of metal determined in the Examples to the value of PEL (Probable effect level) of those metals. Relative toxic unit is the ratio of toxic unit value of each metal to the value of Σ TU in percentage. SPSS was used for statistical analysis. Whether there is a correlation between metal amounts, the strength of this relationship and the direction of the relationship (negative or positive relationship) were examined by correlation analysis. Before performing the correlation analysis, the distribution of the data was examined with the Shapiro Willk test (the Kolmogorov-Smirnov Test was not preferred due to the insufficient

number of data), Pearson correlation tests were applied to the data showing normal distribution, and Spearman correlation tests were applied to the data that did not show normal distribution (Tunca *et al.* 2013).

3. RESULTS AND DISCUSSION

In the study, it was determined that the most Al, Fe and Mn (Al>Fe>Mn>Zn>Ni>Cr>Cu>As>Pb) were accumulated (Table 1). This is an expected result, because the previous studies showed that Fe and Mn comprise most of the natural background of the area in sinkholes (Vesper *et al.*, 2003).

Table 2 demonstrates a comparison of the results obtained and the limit values (SQGs). In concentrations obtained below the ERL (Long and Morgan, 1991) and TEL (Smith *et al.*, 1996) values which are among limit values, no toxic effect is expected for organisms. However, in concentrations obtained above the ERM (Long and Morgan, 1991) and PEL (Smith *et al.*, 1996) values, it is probable to observe a toxic effect (Soliman *et al.*, 2015). As a result, the value of the metal(loid) concentrations tested was below the limit value (Table 2).

Of the metal(loid)s chosen, As, Ni and Pb which are among the elements with a potential of showing a toxic effect, comprised 0.54%. 0.51% and 0.05% of the total toxic effect (1.37), respectively. These values are too low to cause toxic effects for organisms. This condition has also been observed in different studies (Sakan *et al.*, 2020). Examining the contamination factors; all the values were below 1 ($C_f^i < 1$). Concentration values of the metal(loid)s tested, did not pose a problem for pollution.

When EF values are examined; while the Cu (1.74), Ni (2.28) and Pb (2.00) metals had a minor enrichment; As (5.90) had a moderately severe enrichment. The I_{geo} value offers data that may help us understand whether a sediment is polluted or not. Thus, considering the I_{geo} results, it is possible to classify all the metals tested in the study as sediment uncontaminated in terms of As and metals ($I_{geo} < 0$). The I_{geo} results were significantly supported by the EF results. Therefore, it is possible to conclude that the sediment of the Meyil Sinkhole Lake was not

contaminated by the metal(loid)s tested. Evaluating the lake sediment with the m-ERM-Q value; it was at the level of 9% and was found to be moderately impacted owing to the m-PEL-Q value. Thus it is possible to state that the concentrations of elements doing enrichment are probably 9% toxic for organisms and may affect organisms at a moderate level. It has been

reported that the CO₂ activity in carstic systems usually obstructs the water solubility of metals (such as Zn-O-H, Cd-O-H and Pb-O-H) and causes them to precipitate (Vesper *et al.*, 2003). Therefore, the metal assignment in the sinkhole sediment gives us the opportunity of asserting that the lake water there is also poor in metals just like the sediment.

Table 1. Amount of the metal(loid)s tested in the lake sediment (ppm)

	Al	As	Cu	Cr	Fe	Mn	Ni	Pb	Zn
S1	6767.77	9.90	8.29	12.26	5016.38	149.98	16.32	4.01	21.13
S2	8599.20	9.00	11.55	16.02	6620.86	206.75	21.77	5.27	25.40
S3	7049.43	9.10	9.70	13.70	5637.33	189.26	19.08	4.90	24.36
S4	6205.19	8.90	7.77	11.67	4773.91	142.64	15.68	3.78	19.84
S5	7528.05	8.80	9.80	14.48	5914.47	206.44	19.56	4.89	24.42
S6	6777.80	9.30	10.35	13.42	5583.96	191.36	18.87	4.71	23.22
S7	7173.20	9.10	9.70	11.90	5016.50	198.50	19.30	4.90	22.30
S8	7211.10	9.00	8.20	14.40	6019.50	156.70	18.70	5.10	24.50
S9	7089.30	8.90	7.90	13.70	5818.30	200.30	17.90	5.20	26.30
S10	7114.40	9.00	9.90	15.50	5623.60	188.20	16.80	4.70	22.10
Min	6205.19	8.80	7.77	11.67	4773.91	142.64	15.68	3.78	19.84
Max	8599.20	9.90	11.55	16.02	6620.86	206.75	21.77	5.27	26.30
Mean	7151.5	9.1	9.31	13.7	5602.5	183	18.4	4.7	23.4
S.D.	756.13	0.4	1.26	1.43	600.73	25.56	2.04	0.5	1.96

Table 2. Certain SQG values and results of the sediment assessment methods for the lake

	Cu	Pb	Zn	Ni	Mn	Fe	As	Cr	Al
Mean	9.31	4.74	23.36	18.40	183.01	5602.48	9.10	13.71	7151.54
PIN	35.00	50.00	100.00	-	-	-	-	50.00	-
PEL	108.00	112.00	271.00	42.80	-	-	17.00	160.00	-
ERM	270.00	218.00	410.00	51.60	-	-	85.00	370.00	-
TEL	18.70	30.20	124.00	15.90	-	-	5.90	52.30	-
ERL	34.00	46.70	150.00	20.90	-	-	33.00	81.00	-
PERI-RI	5.00	5.00	1.00	-	-	-	10.00	30.00	2.00
Earth crust	45.00	20.00	95.00	68.00	850.00	47200.00	13.00	90.00	80000.00
Contamination factor	0.21	0.24	0.25	0.27	0.22	0.12	0.70	0.15	0.09
Enrichment factor	1.74	2.00	2.07	2.28	1.81	1.00	5.90	1.28	0.75
Geoaccumulation index	-2.86	-2.66	-2.61	-2.47	-2.80	-3.66	-1.10	-3.30	-4.07
Toxic unit	0.05	0.05	0.07	0.51	-	-	0.54	0.15	-
Total toxic unit	1.37								
m-ERM-Q	0.07								
m-PEL-Q	0.14								

Examining some similar studies conducted in carstic systems worldwide; a sediment analysis study conducted in the Lijiang River (China) which is a carstic area, used the mERM-Q for index. The study reported that the Hg, Pb and Zn metals had a medium-low toxicological risk for organisms in the sediment (Xu *et al.*, 2016). A study conducted in a cave in South Africa

evaluated metals (Al, Cd, Cr, Co, Cu, Fe, Pb, Mn, Ni, U and Zn) showing enrichment with human effect in the sediment, via the EF and Igeo. As a result of the study the Cd and U metals were at an extreme contamination level (Preez *et al.*, 2016). Heavy metal enrichments were examined in the Pearl River Basin (a carstic area) and the contamination degree was discussed using the

Igeo index. Cu, Cr and Ni were found to be moderately-highly enriched, while Pb, As and Cd slightly enriched and Zn moderately enriched (Wu *et al.*, 2020).

The mean of the metal and As amounts analyzed from the lake sediment was compared with the published data of other carstic aquatic environments in the world (Table 3). Comparing the results; the Meyil Sinkhole Lake has lower concentrations of metal and As content than other carstic aquatic environments.

A correlation analysis was performed to reveal the metal profile of the Meyil Sinkhole Lake statistically (Table 4). The correlations between

Zn-Pb and Fe are of prime importance. Zn-Pb ($r=0.95$ $p<0.01$), Zn-Fe ($r=0.88$ $p<0.01$) and Pb-Fe ($r=0.87$ $p<0.01$) are the strongest positive correlations in the lake. In addition the correlations between Cr-Al, Fe-Al, Fe-Cr, Mn-Al, Ni-Al, Ni-Mn, Pb-Al and Pb-Mn are very strong. Iron and manganese are commonly present in carstic areas and the presence of these iron and manganese oxides is one of the main reasons why metals enrich (Leveque *et al.*, 2006; Ji *et al.*, 2021). There correlations probably reflect the same or a similar resource input for these metals.

Table 3. Mean metal(loid) contents (ppm) in surface sediments from the Meyil Sinkhole Lake compared with other karst aquatic environments.

Fe	Mn	Al	As	Cd	Cr	Cu	Pb	Zn	Hg	Ni	References
16851	932	6.4	4.8	-	-	17.4	45.7	102.5	-	-	Gutierrez <i>et al.</i> , 2004
17860	-	-	-	4.4	-	188.6	117.4	154.8	-	-	Korfali and Davies, 2005
23500	463	-	-	0.31	42.1	55.1	15.5	58.7	-	31.5	Romic <i>et al.</i> , 2012
26430.7	312.8	131484.4	-	0.1	31.5	12.9	25.9	30.2	-	33.7	Franciskovic-Bilinski <i>et al.</i> , 2014
7.49	0.205	10.23	3.14	1.36	19.6	8.99	24.3	44.8	-	17.4	Duatovic <i>et al.</i> , 2014
-	-	-	26.23	0.61	76.38	43.16	27.84	-	0.45	-	Wu <i>et al.</i> , 2014
-	-	-	23.31	0.53	82.78	45.46	30.22	-	0.27	-	Wu <i>et al.</i> , 2014
-	-	-	5.16	0.25	-	37.43	27.84	-	0.45	-	Yu <i>et al.</i> , 2015
-	-	-	18.05	1.72	56.38	38.07	51.54	142.16	0.18	-	Xu <i>et al.</i> , 2016
5602.481	183	7151.5	9.1	-	13.7	9.3	4.74	23.36	-	18.4	This study

Table 4. Correlation coefficient matrix between the metal(loid)s for lake sediment

	Al	As	Cu	Cr	Fe	Mn	Ni	Pb	Zn
Al	1								
As	-.358	1							
Cu	.474	.254	1						
Cr	.754*	-.322	.582	1					
Fe	.830**	-.383	.353	.851**	1				
Mn	.709*	-.272	.565	.523	.636*	1			
Ni	.770**	-.056	.644*	.474	.636*	.842**	1		
Pb	.775**	-.282	.198	.599	.874**	.732*	.665*	1	
Zn	.632*	-.307	.155	.588	.881**	.699*	.578	.945**	1

*. Correlation is significant at the 0.05 level (2-tailed). **. Correlation is significant at the 0.01 level (2-tailed)

4. CONCLUSION

Metal (loid) accumulation in the sediment of the Meyil Sinkhole Lake was investigated. In addition, the effect of accumulation on the ecosystem was tested with the sediment quality guidelines (SQGs), which includes mathematical formulations. The results show that there is no metal(loid) accumulation in the lake that may be a threat. This study shows that the Meyil Sinkhole Lake is not exposed to metal(loid) pollution caused by anthropogenic activities.

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Şeyda FİKİRDEŞİCİ ERGEN:
Conceptualization, Methodology, Validation, Formal Analysis, Resources, Writing - Original Draft, Writing-Review and Editing, Data Curation, Software, Visualization, Supervision, Project administration, Funding acquisition.

CONFLICT OF INTERESTS

The author declare that for this article they have no actual, potential or perceived conflict of interests.

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