OCCURRENCE OF HEAVY METALS IN THE BUTRINTI LAGOON ECOSYSTEM IN THE SOUTH OF ALBANIA

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ABSTRACT

The Butrinti Lagoon, located in southern Albania, is a vital ecosystem recognized for its rich biodiversity, including the mussel species *Mytilus galloprovincialis*. As a widely consumed bivalve mollusc, the cultivation of this species is crucial, and assessing heavy metal contamination in their tissue is essential for ensuring food safety. This study aims to evaluate the presence of heavy metals in the food chain within the Butrinti Lagoon. Four heavy metalscopper (Cu), chromium (Cr), cadmium (Cd), and lead (Pb), were analyzed in soil, sediment, water, and mussel samples. The findings revealed that heavy metal concentrations in soil samples followed the order $Cr > Cu > Pb >$ Cd, with Cr showing the highest value (378.01 mg/kg). In sediment, the metal concentrations were ranked as Pb > $Cu > Cr > Cd$, with Pb having the highest concentration (64.23 mg/kg). Similarly, in water samples, Pb was the dominant metal (1803 μ g/L), with the order being Pb > Cd > Cu > Cr. Mussel samples also exhibited the highest concentration of Pb, followed by Cr, Cu, and Cd. The consistent presence of Pb at elevated levels across sediment, water, and mussel samples suggests a strong correlation between these matrices, potentially due to the release of Pb from sediments into the water and its subsequent absorption by mussels.

Key words: Heavy metals, *Mytilus galloprovincialis*, Butrinti lagoon, food security.

INTRODUCTION

The contamination of coastal ecosystems is a global issue of significant concern, given the detrimental impact on the aquatic environment. Pollution may be associated with several factors, including industrial sewage, agricultural waste, urbanisation and other human activities (Sherif et. al., 2024). Heavy metals are regarded as pollutants that present a significant ecological risk due to their high toxicity, non-degradability and bioaccumulation. As both essential and non-essential elements involved in a range of biological processes, high levels of heavy metals can have adverse effects on human health (Rozirwan et. al., 2024). Heavy metals that are introduced into aquatic ecosystems from a variety of sources will inevitably settle at the bottom of the water column and accumulate in the biota (Bhuyan et. al., 2023). Sediments were identified as the primary reservoir for heavy metal pollutants, which represent the predominant habitat for biota, particularly in benthic species groups (mollusca, crustaceans, and polychaeta), fish, and shrimp (Pandiyan et. al., 2021; Rozirwan et. al., 2024). Heavy metals can exert direct effects on organisms by accumulating in the body or indirect effects by migrating up the food chain to the next trophic

level. In the food chain, heavy metals are accumulated from two sources: bioaccumulation from the food source and accumulation from the environment, such as water or sediment (Amany 2024; Ali, et. al., 2021).

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The Butrinti lagoon (16 km²) is one of the most intriguing lagoons of tectonic origin in the southern Albanian region and is the most intensively utilised in the aquaculture of the mussel (*Mytilus galloprovincialis*) (Topi et al., 2013). The lagoon is subject to pressure from several human activities, including agriculture, tourism and urban development. Mussel farming represents a significant economic activity in the region. These activities are responsible for the increase in heavy metal concentrations within this ecosystem. In recent years, elevated levels of heavy metals have been reported in this lagoon (NEA, 2023). It is therefore important to accurately measure the levels of heavy metals and other nutrients in aquatic environments to control and monitor pollution levels (Koto et. al. 2023). The aim of this study was to evaluate the concentrations of heavy metals in the soils around the lagoon, sediment, water and mussel samples, as this provides a key indicator of food security. Furthermore, a comparative evaluation with other studies will provide insights into the changes that have occurred in this lagoon and help identify the source of pollution in this ecosystem.

MATERIAL AND METHODS

The study area

The study was realized in Butrinti lagoon. Butrinti lagoon with coordinates [39°47′0″N](https://geohack.toolforge.org/geohack.php?pagename=Liqeni_i_Butrintit¶ms=39_47_0_N_20_2_0_E_type:waterbody) 20°12′0″E, has an area of 1600 ha, it has a maximum 7.1 km length and 3.3 km wide. The maximum depth is 21.4 m with an average of 11 m. Butrinti lagoon lies within the Mediterranean Climatic Zone, Central Sub-zone (Sulçe et. al., 2011). This lagoon has tectonic origin, and from hydrographic regimen is typical coastal.

Sample collection and preparation

In May 2024, five sampling points (B1, B2, B3, B4, B5) were identified within the lagoon, and water samples were collected from each point in 0.5 L polyethylene bottles (Figure 1). The samples were collected from a depth of 50 cm below the water surface and preserved by adding nitric acid to achieve a pH of less than 2, specifically 0.5 ml of concentrated acid was added per 100 ml of sample. The water samples were then analyzed for heavy metals, including copper (Cu), cadmium (Cd), chromium (Cr), and lead (Pb), using atomic absorption spectrometry with a graphite furnace (ISO 15586:2003). The results are expressed as the mass of the analyte (in micrograms, µg) per litre of water.

Figure 1. Water sampling points in Butrinti lagoon.

At representative sampling points, five soil samples (TB1, TB2, TB3, TB4, TB5) and three sediment samples (SB2, SB3, and SB4) were collected from a depth of 0-30 cm (Figure 2).

Figure 2. Sampling points of soil and sediment samples in Butrinti lagoon

The soil and sediment samples were collected from the same locations as the water samples. The samples were then air-dried, sieved using a 2mm sieve, and the fraction of samples with a diameter of less than 2 mm were used for further analysis. The total concentrations of Cd, Cr, Cu and Pb were determined in soil and sediment samples using the following procedure: A total of 0.3 g of soil and/or sediment samples were combined with 8 ml of concentrated nitric acid (HNO₃) and 2 ml of 33% hydrogen peroxide (H₂O₂). The resulting mixture was then subjected to microwave-assisted digestion at 180°C for 25 minutes. The concentrations of heavy metals were subsequently determined by atomic absorption spectrometry (AAS) (USEPA 1997).

Three sampling points (1, 2, 3) were selected for the collection of mussels, situated in the north, western and southern regions of the lagoon, respectively (Figure 3).

Figure 3. Sampling points for mussels in Butrinti lagoon

At each sampling point, three samples of mussels were collected, resulting in a total of nine samples. To extract heavy metals from the mussels, 0.3 g of dried tissue was placed in a container with 8 ml of HNO₃ and 2 ml of H₂O₂. The samples were then mineralised using a microwave digester. Following this, the dissolved sample was transferred to a 50 ml balloon, where it was adjusted to distilled water. The concentrations of heavy metals were determined by graphite furnace atomic absorption spectroscopy (AAS) (USEPA, 1997). All the analyses were performed in the Laboratory of the Department of Environment and Natural Resources at the Agricultural University of Tirana.

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Assessment of heavy metal contamination

The degree of water contamination with heavy metals was assessed in accordance with the prescribed limit values for surface water as outlined in Directives 75/440/EEC and 2008/105/EC. To assess the ecological impact of sediments contaminated with heavy metals, the guidelines outlined in (NOAA-USA, 1995) were followed. Similarly, the (EC) No 1881/2006 reviewed in 2023 standard on maximum levels for certain contaminants in food was used to assess the concentration of heavy metals in mussel samples. To evaluate the origin of heavy metals in this ecosystem, Pearson correlation analysis was performed using the SPSS 20 software.

RESULTS AND DISCUSSION

Heavy metals in soil and sediment samples

The results for heavy metals in soil and sediments samples are presented in Table 1. The mean concentrations of heavy metals in soil showed a decreasing trend, with chromium (Cr) having the highest concentration, followed by copper (Cu), lead (Pb), and then cadmium (Cd). Chromium is a multifaceted metal that is widely distributed throughout the Earth's crust in a range of forms, including those found in plant and ore deposits. Its presence can be attributed to both natural sources and human activities, including industrial processes such as ship breaking, stainless steel production, and plating (Bhuyan et. al., 2023; Maurya and Kumari 2021).

The concentration of *chromium (Cr)* in soil samples ranged from 38.23 mg/kg (TB5) to 532.49 mg/kg (TB1), with an average value of 378.01 mg/kg. A comparison of these values with a previous study by (Topi et al., 2013) on the soil of this lagoon reveals that they are considerably higher, as well as exceeding the average concentration of chromium in Albanian soils (150 mg Cr/kg) (Gjoka et. al., 2010). Moreover, these concentrations exceed the World Health Organization's (WHO) permissible limit for chromium in soil, which is set at 100 mg/kg (WHO, 1972).

Table 1. The values of heavy metals in soil and sediments in Butrinti lagoon.

Copper (Cu), a metal that is released into the environment through various avenues, including mining, metal processing, agriculture, and chemical industries, is employed widely in both industrial and agricultural practices (Yunus, et. al., 2020). The concentration of Cu in the soil of the Butrinti lagoon ranges from 58.29 mg/kg (TB2) to 83.68 mg/kg (TB1). These values are similar to those reported in the study of (Topi et al. 2013) but exceed the limit set by the WHO (36 mg/kg). *Lead (Pb)* is a stable element that presents a significant risk to human and animal health, particularly affecting the kidneys and nervous systems. In soil samples, lead concentrations ranged from 25.14 mg/kg (TB5) to 91.74 mg/kg (TB4). The mean values and most of the samples indicate that lead (Pb) levels are within the WHO's permissible limit for heavy metals in soil, set at 85 mg/kg. However, these values are considerably higher than those reported in the 2013 study (Topi, et. al., 2013) on heavy metals in soil from the Butrinti lagoon.

The concentration of *cadmium (Cd)* in soil samples ranged from 0.34 mg/kg (TB4) to 2.29 mg/kg (TB1). Additionally, these values are deemed to be within the range of those observed in the previous studies (Topi, et. al., 2012; 2013), but higher average concentrations than the WHO limit (0.8 mg/kg). A comparison with the EU standard (86 / 278 / EEC, 1986) reveals that the concentrations of these heavy metals are within the specified limit values. The soil in the vicinity of the Butrinti lagoon is not subject to industrial activities such as mining and smelting, nor is it affected by energy-intensive industries that could potentially lead to an increase in heavy metal concentrations. It is possible that the high levels of heavy metals (HMs) are naturally occurring, given that they are derived from the weathering of minerals in the Earth's crust. Heavy metals (HMs) in rocks can be released into the soil through various natural processes, such as erosion, leaching, biological activity, terrestrial processes, and surface winds. Studies also report that in the Mediterranean region, autumn and spring rainstorms saturate the soil surface with water, facilitating the release of cations that are not chemically bound to clay (Topi et. al., 2012). Thus, in this context the heavy metals in soil samples are expected to be detected in the high concentrations.

In sediment samples, the concentration of heavy metals was found to decrease in the following order: $Pb > Cu > Cr$ $>$ Cd. The concentration of lead (Pb) in sediments ranged from 24.16 mg/kg (SB4) to 140.64 SB3. These concentrations fall within the range of ERL and ERM, respectively, at 46.7-218 mg/kg. Additionally, the concentration of lead is higher than the mean value of the same element in the previous study (Topi et. al.,2012), where Pb was 31.2 mg/kg. The concentration of copper (Cu) ranged from 48.85 mg/kg (SB2) to 68.51 mg/kg (SB4).The values of Cu are within the interval of ERL and ERM of biological effects, but two times higher than the concentration observed in the former study (Topi et. al., 2012). The chromium (Cr) range is from 13.16 mg/kg (SB2) to 52.63 mg/kg (SB4). A comparison with sediment quality guidelines reveals that the values of Cr are lower than ERL (effect range lower) and lower than the concentration of Cr definitively established in the sediments during the 2012 study (Topi et. al., 2012), where the average value was 56.5 mg/kg. The concentration of cadmium (Cd) in the samples ranged from 0.46 mg/kg (SB3) to 1.32 mg/kg (SB2). The mean concentration of Cd is 0.84 mg/kg and is higher than that reported in the 2012 study (Topi et. al., 2012), which was 0.125mg/kg and within ERL values for the indication of heavy metals in different aquatic organisms. The results indicate that the elevated concentrations of heavy metals in the sediments, which serve as the final sink for these contaminants, originate from urban sources or fertilizer application. The concentrations of Cd and Pb were higher than in previous studies (Topi et. al., 2012; 2013), likely due to the intensified agricultural activities.

The correlation of heavy metals in soil and sediment samples

In table 2 are presented the Pearson correlation coefficients of heavy metals between soil and sediment samples.

Table 2. The correlation between soil and sediment samples for heavy metals

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

The correlation coefficients demonstrate that all soil samples are significantly correlated with all heavy metals at the 95% and 99% level. Notably, TB5 is the one soil sample that does not exhibit correlation with the others. However, TB5 is positively correlated with coefficient (971*) with sediment sample, SB4. This indicates that, except for TB5, the heavy metals present in the soil around Butrinti lagoon have a similar origin. Most of the heavy metals in the soil samples may have originated from a variety of anthropogenic activities, including discharge from the urban area of Ksamil and natural weathering of rocks. However, in TB5, which is an agricultural soil, as well as in the sediments of this ecosystem, the origin of the heavy metals is likely due to agricultural activities.

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Heavy metals in the surface water of Butrinti lagoon

The concentrations of heavy metals in the water samples are presented in Table 2. In descending order of concentration, the elements were $Pb > Cd > Cu > Cr$. The concentration of *lead (Pb)* in the water samples ranged from 1081.8 μ g/l (B5) to 1802.6 μ g/l (B3). These values are considerably higher than the EU standard, resulting in the classification of these waters as class V with a Pb concentration exceeding 2.5 μ g/l.

Furthermore, the mean Pb concentration was higher than that reported in the previously mentioned studies (Topi et. al., 2012; 2013) and the National Environment Agency (NEA, 2023) report where the mean Pb concentration in water was 618 μ g/L. The concentration of *cadmium (Cd)* in the water samples ranged from 0.45 μ g/l (B3) to 10.06 µg/l (B5). Similarly, the concentration of Cd is relatively higher, classifying the water of the lagoon mostly as class V ($>$ 5 µg/l). It is notable that the values observed in the present study are higher than those reported in the 2012 and 2013 studies (Topi et. al., 2012; 2013). However, when these values are compared with the mean values reported by NEA in 2023 (Cd, 21 µg/l), they are found to be lower.

Copper (Cu) is a highly prevalent element that occurs in its natural state. Even at concentrations as low as 0.0001 mg/L, copper can have toxic effects. It is established that copper causes brain damage in mammals (Korbla Doamekpor, 2017).

The data presented in Table 3 demonstrate that the concentrations of copper detected in the water samples were relatively low, falling below the EU standard and classifying the water as class I (i.e., $\langle 50 \mu g/L \rangle$). Moreover, these levels were found to be lower than those reported in the 2013 study (average values of 10.26 µg/l) (Topi et. al., 2013) and in line with the level reported by the NEA 2023. Although copper compounds are used in fungicides, algicides, insecticides, and fertilizers in soils, the lower concentration of Cu in water can be explained by its mobility and the tendency to bind to sediments (Topi et. al., 2012).

Sampling station	$Cu \mu g/l$	$Cr \mu g/l$	$Cd \mu g/l$	Pb μ g/l	
B1	0.73	0,32	8,19	1322,5	
B ₂	0,6	0,05	8,95	1113,6	
B ₃	0,31	0,04	0,45	1802,6	
B4	0,46	0,042	4,11	1598,8	
B ₅	3,28	0,041	10,06	1081,8	
Average	$1,076 \pm 1,24$	0.10 ± 0.12	$6,35\pm3,99$	1383,86±312,08	
EU Guideline	50	50		50	

Table 3. The concentrations of heavy metals in the water samples of Butrinti lagoon

The results indicated that the concentration of *chromium (Cr)* ranged from 0.04 µg/l (B3) to 0.32 µg/l (B1). The mean concentration was found to be lower than that reported in the 2012 study (mean value 1.63 μ g/l). The higher levels of Pb and Cd in the lagoon can be attributed to anthropogenic inputs into the water, as well as weathering and leaching from agricultural lands.

Notwithstanding the fact that these heavy metals in sediments were within the standard limit, their concentration in water was considerable. This indicate that heavy metals may be exchanged within the water column due to changes in physicochemical conditions. Alternatively, it can be argued that the lead in water originates from urban discharges in surrounding areas before being deposited in sediment sinks (Topi et. al., 2012).

Heavy metals in mussels' samples in Butrinti lagoon

In accordance with the EU standard on maximum levels for specific contaminants in food and the repeal of Regulation (EC) No 1881/2006, the most pertinent interest is the limit value for heavy metals such as Hg, Pb and Cd, which are the most toxic heavy metals present in food. In this study, the focus was on lead, cadmium, chromium, and copper. The concentration of these elements was found to decrease in the following order: $Pb > Cr >$ $Cu > Cd$ (Table 4).

Sampling station	Cd	Cr	Cu	Pb
Unit	mg/kg	mg/kg	mg/kg	mg/kg
$W1-1$	1,97	1,96	4,24	3,9
$W1-2$	1,99	2,37	3,15	3,59
$W1-3$	1,97	2,85	2,95	3,56
$S2-1$	2,47	6,64	2,3	4,02
$S2-2$	2,47	7,19	3,67	4,52
$S2-3$	2,28	7,04	3,48	4,43
$N3-1$	2,25	0.91	3,16	4,78
$N3-2$	2,31	0,92	1,3	4,86
$N3-3$	2,24	0,86	2,24	4,86
Average	$2,22\pm0,19$	$3,42\pm2,74$	2.94 ± 0.88	$4,28 \pm 0,52$
(EC) No 1881/2006 and FAO*		$0.5*$	٠	1.5

Table 4. The concentration of heavy metals in mussel samples.

It seems probable that the situation in the lagoon is similar regarding the results of heavy metals in mussels, based in their concentrations. The concentration of *lead (Pb)* ranged from 3.56 mg/kg (W1-3) to 4.86 mg/kg (N3-2; N3-3). This concentration exceeds the EU standard (EU 2023/915; 1881/2006) (Pb= 1.5 mg/kg) and is also higher than the average level reported by Topi et. al, 2013 (Pb= 0.219 mg/kg). The *chromium* concentration (Cr) ranged from 0.86 mg/kg (N3-3) to 7.19 mg/kg (S2-2). In accordance with the FAO and WHO 1972, the permissible limit for Cr is 0.5 mg/kg. Therefore, the concentration of Cr in the mussel samples from this study is above the permitted threshold.

Additionally, the concentrations of *copper (Cu)* and *cadmium (Cd)* have been specified. The concentration of copper ranges from 1.3 mg/kg (N3-2) to 4.24 mg/kg (W1-1), while that of cadmium varies from 1.97 mg/kg (W1-1) to 2.47 mg/kg (S2-1; S2-2). In accordance with the EU standard (EU 2023/915; 1881/2006), the permitted level of Cd, a toxic metal, is 1 mg/kg. However, the samples from this study shown a higher concentration of Cd. Considering these findings, it is evident that there is a potential risk associated with the consumption of mussels from the Butrinti lagoon. The accumulation of heavy metals in these mussels could potentially be transferred into the human body, necessitating further investigation.

The correlation of heavy metals in water and mussels' samples

Table 5 presents the correlation coefficients between heavy metal concentrations in water and mussel samples. Similarly, the correlation coefficients indicate a similar relationship between the variables in soil and sediments. The correlation between water samples was perfect, with a significance of 100% and a coefficient of 1. This indicates that the source of the heavy metals in the water samples is consistent across all water ecosystems. The level of these elements may be attributed to several factors, including anthropogenic activities, different discharges, wastewater from the surrounding area, fertilizers, or the release of heavy metals from sediments. As previously observed, the concentration of heavy metals in sediment samples are within the limit standards.

Therefore, it can be postulated that the alterations in physicochemical parameters within the sediments may have contributed to the presence of heavy metals in the water samples. Nevertheless, a similar phenomenon is observed in the mussel samples, where the concentration of heavy metals exceeds the established standard. Sediments are a common source of metals in mussel populations. In uncontaminated sites, metal concentrations in sediments originate from geological materials (Sanz-Pradaa et. al., 2022). Thus, our mussel populations exceed the amount of heavy metals due to the absorption from sediments and waters too. These heavy metals can be mostly from geological materials of surrounding areas.

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		$W1-1$	$W1-2$	$W1-3$	$S2-1$	$S2-2$	$S2-3$	$N3-1$	$N3-2$	$N3-3$	B1	B ₂	B3	B4	B5
W1-1	Pearson Correlation		.910	.695	-441	$-.233$	$-.225$.799	.382	.615	.479	.478	.481	.480	.480
W1-2	Pearson Correlation	.910		.915	-112	.048	,062	.831	.585	.739	.745	.744	.747	.746	.745
$W1-3$	Pearson Correlation	.695	.915		,299	.436	,450	.603	.491	.573	.737	,736	.740	.739	,736
$S2-1$	Pearson Correlation	-441	$-.112$.299		.960°	.961	-483	$-.176$	-339	.051	.050	.054	.053	.048
$S2-2$	Pearson Correlation	.233	.048	.436	$.960*$		$.000**$	-433	$-.267$	$-.360$.015	,014	.019	.017	,012
$S2-3$	Pearson Correlation	.225	.062	,450	$.961*$	$1.000**$		-416	$-.246$	$-.340$.036	,035	.040	.039	,034
N3-1	Pearson Correlation	.799	.831	.603	-483	$-.433$	-416		.846	$.960*$.823	,823	.823	.823	.824
$N3-2$	Pearson Correlation	.382	.585	.491	$-.176$	$-.267$	$-.246$.846		$.961*$.946	.946	.944	.945	.946
N3-3	Pearson Correlation	.615	.739	.573	-339	$-.360$	$-.340$	$.960*$	$.961*$.922	.923	.921	.922	.923
B1	Pearson Correlation	.479	.745	,737	.051	,015	,036	,823	.946	,922		$1.000**$	$1.000**$	$1,000**$	1.000^{**}
B ₂	Pearson Correlation	.478	.744	.736	.050	.014	,035	.823	.946	.923	$1,000**$		$1.000**$	$1,000**$	$1,000**$
B ₃	Pearson Correlation	.481	.747	.740	.054	.019	.040	.823	.944	.921	$1.000**$	$1.000**$		$1.000**$	$1.000**$
B ₄	Pearson Correlation	.480	.746	.739	.053	,017	,039	,823	.945	,922	$1,000**$	$1.000**$	$1.000**$		$1.000**$
B ₅	Pearson Correlation	.480	745	736	.048	.012	.034	.824	.946	.923	$1.000**$	0.00^{**}	$1,000**$	$1,000**$	

Table 5. The correlations coefficients in water and mussel samples

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

CONCLUSIONS

- This study provides a comprehensive overview of the status of heavy metal contamination in the Butrinti Lagoon, focusing on their presence in the food chain, which is critical for assessing food security. The concentration of heavy metals in soil samples followed a descending order, with chromium (Cr) having the highest levels, followed by copper (Cu), lead (Pb), and cadmium (Cd). These concentrations were higher than those reported in previous studies and exceeded standard limits. In sediment samples, the heavy metal concentrations ranked as $Pb > Cu > Cr > Cd$, and their levels were within ERL and ERM values for biological effect.
- The higher concentrations of heavy metals in sediment, was reflected also in water samples, particularly for Pb and Cd. A similar pattern was observed in mussel samples, where heavy metal concentrations exceeded both EU standards and those reported in previous studies. The variability in heavy metal levels across different components of the ecosystem is likely due to fluctuations in physic-chemical conditions. This situation is highlighted by the correlation analysis between soil and sediment samples, as well as between water and mussel samples.
- Based on the evidence, it is concluded that while the heavy metals in the Butrinti Lagoon predominantly originate from natural sources, anthropogenic activities, such as water discharge, weathering and leaching from agricultural lands, and pesticide use, contribute significantly to the increase in heavy metal concentrations. Importantly, there is a potential health risk associated with the consumption of mussels from the lagoon, as the bioaccumulation of heavy metals in mussels could be transferred to humans through the food chain.

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