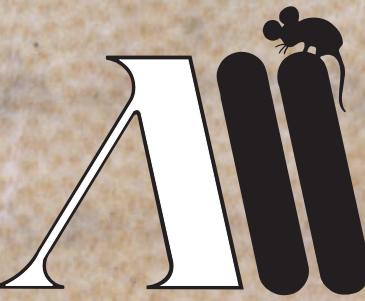


# ANNALES



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Series Historia Naturalis, 31, 2021, 2*



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## HEAVY METAL CONCENTRATIONS IN TISSUES OF RED MULLET, *MULLUS BARBATUS* (MULLIDAE) FROM THE SYRIAN COAST (EASTERN MEDITERRANEAN SEA)

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### *ABSTRACT*

*Cadmium, lead and copper levels were measured in muscles and liver of red mullet *Mullus barbatus* Linnaeus, 1758 caught off the Syrian coast between August 2019 and May 2020. It was found that the metals were accumulated in different tissues of *M. barbatus* by various levels, where the non-edible parts accumulated more metals than the edible muscles. The highest average levels of lead ( $0.164 \pm 0.098 \mu\text{g/g}$ ), and copper ( $8.69 \pm 2.75 \mu\text{g/g}$  wet weight) were recorded in the liver. The concentrations of Cd, Pb, and Cu measured in edible muscle flesh were lower than the maximum acceptable limit set by FAO/WHO for human consumption. The order of the metal concentrations found in *M. barbatus* was Cu> Pb> Cd. Changes in metals concentrations in the tissues of *M. barbatus* were observed versus areas and seasons.*

**Key words:** Syria, heavy metals, *Mullus barbatus*, bioaccumulation, eastern Mediterranean Sea

## CONCENTRAZIONI DI METALLI PESANTI IN TESSUTI DI TRIGLIA DI FANGO, *MULLUS BARBATUS* (MULLIDAE) LUNGO LA COSTA SIRIANA (MEDITERRANEO ORIENTALE)

### *SINTESI*

*I livelli di cadmio, piombo e rame sono stati misurati nei muscoli e nel fegato della triglia di fango *Mullus barbatus* Linnaeus, 1758, catturata al largo della costa siriana tra agosto 2019 e maggio 2020. I metalli sono stati accumulati in diversi tessuti di *M. barbatus* in concentrazioni varie, e le parti non commestibili hanno accumulato più metalli che i muscoli commestibili. I livelli medi più alti di piombo ( $0,164 \pm 0,098 \mu\text{g/g}$ ) e di rame ( $8,69 \pm 2,75 \mu\text{g/g}$  peso umido) sono stati registrati nel fegato. Le concentrazioni di Cd, Pb e Cu misurate nella parte muscolare commestibile erano inferiori al limite massimo stabilito dalla FAO/OMS per il consumo umano. L'ordine delle concentrazioni dei metalli trovato in *M. barbatus* era Cu> Pb> Cd. I cambiamenti nelle concentrazioni di metalli nei tessuti di *M. barbatus* sono stati osservati rispetto alle aree e alle stagioni.*

**Parole chiave:** Siria, metalli pesanti, *Mullus barbatus*, bioaccumulo, Mediterraneo orientale

## INTRODUCTION

Five species are reported to date in the Syrian waters, two are indigenous species such as red mullet, *Mullus barbatus* Linnaeus, 1758 and striped red mullet *M. surmuletus* Linnaeus, 1758 (Saad, 2005; Ali, 2018). Three are alien species incoming from the Red Sea through Suez Canal into the Mediterranean Sea, as Lessepsian migrants (*sensu* Por, 1978), for instance golden-banded goatfish *Upeneus moluccensis* (Bleeker, 1855), Por's goatfish *U. pori* Ben-Tuvia & Golani, 1989 and *Parupeneus forsskali* (Fourmanoir & Guézé, 1976). These three alien species are at present harvested in large quantities and sometimes more than the 2 indigenous mullid species, the best instance being *U. moluccensis* (Saad et al., 2017).

Mullid species constitute 3.1% of the number of bony fishes collected in the Syrian marine waters, and 8.9% of the total catch by artisanal fishing gears (Ullman et al., 2015; Saad et al., 2017). Mullid species display a large economic value in the area because they are locally very appreciated for human consumption, and among them mainly *M. barbatus* (Saad & Sabour, 1998). This species is known as a bottom feeding carnivorous species at the top of the food chain (Saad & Sabour, 1998), and therefore could be expected that bio-accumulation levels of heavy metals rise.

A bioaccumulation of heavy metals in the different fish tissues has been previously studied (Saad & Hammoud, 2007; Mohamed, 2008; Turan et al., 2009; Abdallah, 2013; Aytekin et al., 2019), also in *M. barbatus* showing that the species tissues accumulated high concentrations of heavy metals (Sunlu, 2004; Benedicto et al., 2007; Dural et al., 2010; Findik & Çiçek, 2011; Allan et al., 2016). However, similar investigations have not been carried out for specimens of *M. barbatus* from the Syrian waters which constitute the aims of the present papers in order to preserve human health of risk assessment.

The purpose of the present study consists to assess selected metal (Cu, Pb, and Cd) concentrations in muscles and liver of *M. barbatus* caught by commercial fisheries from three areas located on the coast of Syria.

## MATERIAL AND METHODS

### Study area

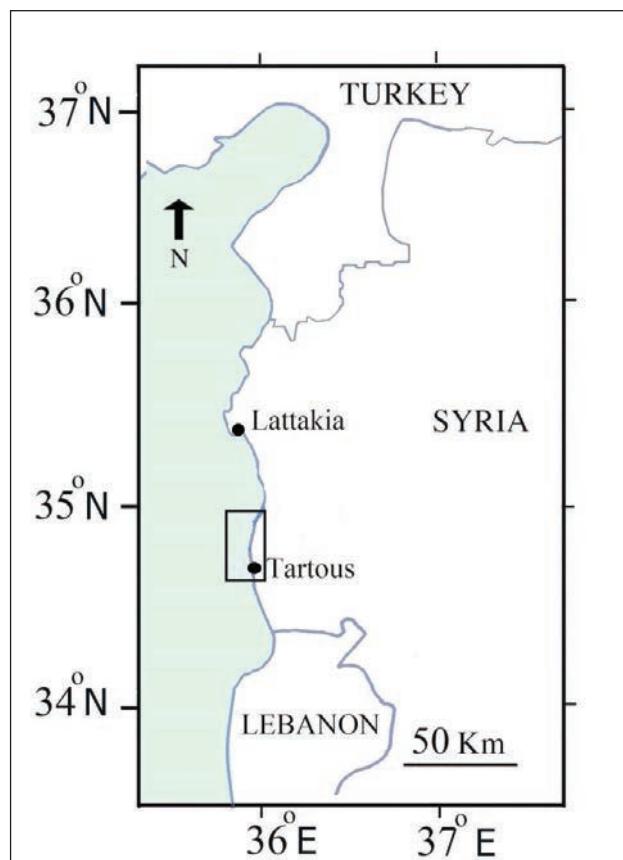
The sampling stations were selected based on main factors such as industrial effluents and sewageare being discharged into the waters of Syrian coast (Fig. 1). Therefore, three different sites were chosen. The first site (T1) was relatively close to in-

dustrial pollution sources ( $34^{\circ}59'46''$  N,  $35^{\circ}53'21''$  E). The second site (T2) was performed based on its relation with the thermic power station activities ( $35^{\circ}10'11''$  N,  $35^{\circ}55'36''$  E). The third site (T3) represents an area for the sewage downstream ( $34^{\circ}53'09''$  N,  $35^{\circ}52'57''$  E).

### Sample preparation and analysis

A total of 12 specimens of *M. barbatus* were studied, they were captured commercial bottom-trawler or gill net fisheries from August 2019 to May 2020, at the 3 stations (Tab. 1). Total length (TL) was recorded to the nearest mm and total body weight (TBW) to the nearest 0.1 gram. TL ranged between 92 and 170 mm and TBW between 20.9 and 60 g.

The collected specimens were preserved in plastic boxes filled with ice, and then delivered at the laboratory. They were washed with distilled water, dried in filter paper and stored at -25°C until dissection. At about 3 gram sample of fish muscles



**Fig. 1: Map of the Syrian coast with rectangle indicating the sampling area of *Mullus barbatus*.**  
**Sl. 1: Zemljevid sirske obale s pravokotnikom, ki prikazuje vzorčevalno postajo ulova bradačev.**

**Tab. 1: Minimum, maximum and mean metal concentrations in the tissues of the red mullet, *Mullus barbatus*, from the coastal waters of Syria and comparison of different sites (µg/g wet wt).****Tab. 1: Minimalne, maksimalne in srednje vrednosti koncentracij težkih kovin v primerih bradača, *Mullus barbatus*, ujetih v obalnih vodah Sirije in primerjava med različnimi lokalitetami (µg/g mokre teže).**

Tissue	Site	Mean	Cu Range	Mean	Pb Range	Mean	Cd Range
Muscles	T1	0.462	0.35-0.577 a	0.0281	0.018-0.038 a	0.0062	0.006 - 0.007 a
	T2	0.619	0.056-0.65 b	0.0651	0.05-0.074 b	0.0205	0.019-0.020 c
	T3	0.578	0.52-0.62 ab	0.0778	0.067-0.09 c	0.0172	0.015-0.018 b
Liver	T1	7.85	5.3-12.24 a	0.0497	0.031-0.07 a	-	-
	T2	8.91	5.69-12 a	0.193	0.18-0.21 b	-	-
		9.91	7.75-14 b	0.250	0.23-0.27 c	-	-

Letters a, b and c show differences among sites. Means with the same letter are not statistically significant, p>0.05.

and part of liver, were removed. The wet digestion method was used in the analysis of the heavy metals (Saad & Hammoud, 2007; Abdallah, 2013; Turan et al., 2019).

Samples were transferred into digestion flasks and treated with 5 ml HNO<sub>3</sub> (ultrapure, Merck) on the hot plate until the color turns into light yellow, nearly white. After this process the samples were transferred to 25 ml flanks and added double distilled water until 25 ml. The solution was filtered by filter papers.

At each step of the digestion processes, acid blanks (laboratory blank) were prepared using identical procedure to ensure that the samples and chemicals used were not contaminated from any of the mentioned possible sources. They contain the same digestion reagents as the real samples with the same acid ratios but without the fish sample. They were analyzed by Atomic Absorption Spectrophotometer (Shimadzo-AA6800) before the real samples, to check if it will give the exact values of heavy metals in real samples.

### Statistical analysis

Statistical differences between mean metal concentrations in different sites were evaluated using one way ANOVA. The difference between the seasons was analyzed through Student t-test.

## RESULTS AND DISCUSSION

### Heavy metals

Cu is the most abundant of metals examined (Tab. 1), it is an essential element since it plays important roles in biological systems (Kayhan et al., 2017). Conversely, lead and cadmium have caused harmful and toxic effects for human health even in trace

amounts (Tepe et al., 2008). Copper concentrations ranged from 0.35 to 0.65 mg/g wet weight (ww) in muscle, the highest level occurring at station site T2, and from 5.30 to 14.00 mg/g ww in liver, with the highest level at site T3.

Copper concentrations vary significantly (p<0.05) in the muscle tissues, although copper concentrations in liver tissues did not show significant differences between T1 and T2 (p>0.05) (Tab. 1). Pb and Cd belong to non-essential, do not any function in biochemical processes.

This study shows that there were little variations in mean concentration of Cd and Pb in all sites investigated. In this study the mean Pb levels in muscle and liver were 0.0569 µg/g and 0.164 µg/g respectively (Tab. 1). Pb levels both in muscle and liver tissues showed significant differences between region. Mean Cd levels in the muscles of red mullet from T1, T2 and T3 from Syria were 0.0062, and 0.0172 mg/g, respectively. Cd levels in muscle tissues were statistically different between region. Pb and Cd concentrations were found higher in T3 and T2 station respectively. Our results show that metal accumulation is lowest in muscles, while it is high in liver in all sites. This probably due to their physiological roles in fish metabolism. Dural et al. (2010) determined that large amount of metallothione in induction occurs in the liver tissues.

Some authors have addressed measurements of Cu, Pb and Cd in fish from different regions of the world (Tab. 2). It appears differences between metal concentration in this study and those of previous studies. Copper concentrations in liver tissues found in this study were higher than those from Çandarlı Bay. (Tas et al., 2011), but displayed Copper concentrations in muscles tissues similar values than those of reported from Izmir Bay and Çandarlı Bay by Sunlu (2004) in Table 2. Cd concentrations have been less studied when than other metals in *M. barbatus* (Sunlu, 2004).

**Tab. 2: Levels of heavy metals in *Mullus barbatus* from different areas of the world (µg/g wet weight)**  
**Tab. 2: Vsebnosti težkih kovin pri vrsti *Mullus barbatus* iz različnih predelov sveta (µg/g mokre teže).**

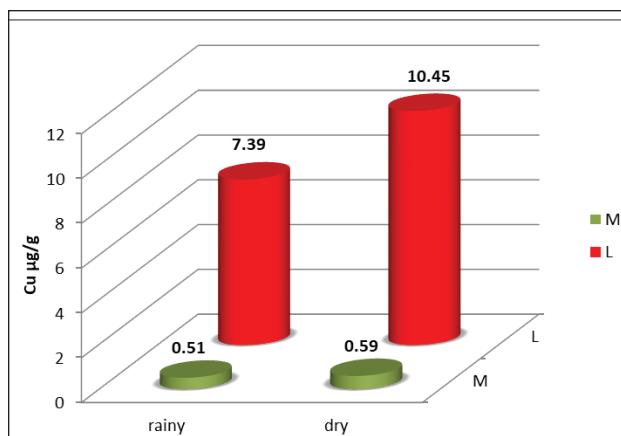
Tissue	Authors	Area	Cu	Pb	Cd
Muscles	Tas et al. (2011)	Çandarli Bay	0.11-1.25	1.20-9.74	-
	Sunlu (2004)	Izmir Bay	0.11-0.50	0.80-2.60	-
	Turan et al. (2009)	Black Sea	0.77-1.24	0.14-0.82	0.06-0.29
	Fındık & Çiçek (2011)	Black Sea	4.05	1.05	1.01
Liver	Tas et al. (2011)	Çanarli Bay	0.62 -2.09	5.30-12.52	-
	Mariji & Raspor (2007)	Eastern Adriatic	0.15-0.68	-	-
	Tepe et al. (2008)	Turkish seas	1.11-26.7	0.66-5.20	-

### Seasonal variations in concentrations of metals in *M. barbatus*

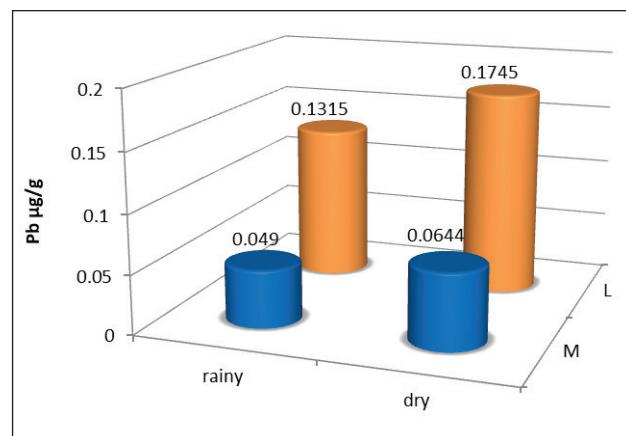
Copper contents in specimens versus seasons were different (Fig. 3). The mean Cu concentration in *M. barbatus* muscles from Syrian coast ranged between 0.51 / µg /g in rainy season and 0.59 mg/g in dry season and 7.39 to 10.45 at dry µg/g in liver (Fig. 2), and these values were significantly different ( $t$ -value = -3.07 , -4.80 ;  $p$  = 0.005;  $df$  = 1).

The mean Pb concentration in liver tissues did not show significant differences between season (Fig. 2). Conversely, the mean concentration of Pb in muscle of *M. barbatus* ranged from 0.049 mg/g to 0.0644 mg/g and show significant differences between seasons (Fig.3) ( $t$ -value = -4.34.;  $p$  = 0.007).

The mean Cd concentrations ranged from 0.006 at rainy to 0.022 at dry mg/g in muscle (Fig. 4). Cd levels in muscle tissues were not different between seasons ( $t$ -value = -1.66;  $p$  = 0.158). There was a significant increase in accumulation of Cu, Pb, and Cd in tow tissues of *M. barbatus* from Syrian coast seasonally. The accumulation of metals in white muscle, and liver increased in dry season than rainy season this may be related to increase human activities in this seasons and increase in physiological activity of fish due to increase of temperature this confirmed by previous studies such as (Jakimska et al., 2011). Similar increases in metal levels were observed during summer in fish species from Iskenderun Gulf (Aytekin et al., 2019).



**Fig 2: Differences of Cu concentrations in *Mullus barbatus* among the seasons: M = muscles, L = Liver.**  
**Sl. 2: Razlike v koncentraciji Cu pri vrsti *Mullus barbatus* v različnih sezonah: M = mišice, L = jetra.**



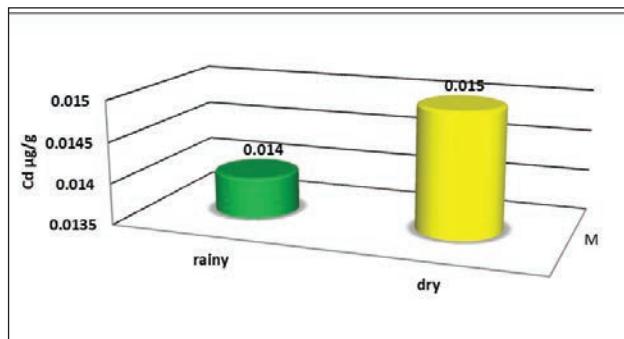
**Fig 3: Differences of Pb concentrations in *Mullus barbatus* versus seasons.**  
**Sl. 3: Razlike v koncentraciji Pb pri vrsti *Mullus barbatus* v različnih sezonah.**

### Human health risk assessment

Fish species are considered as one of the main protein sources of food for human health, because of having rich contents of essential minerals, vitamins and unsaturated fatty acids (Zaza et al., 2015). In this study, the measured metal concentration in edible tissues, such as of *M. barbatus* were compared with some existing standards for human consumption.

From Marmara Sea, the levels of Cd and Pb were found as very high according to tolerance limits of the World Health Organization (WHO, 1996) standards (Kayhan et al., 2017). For the fish samples from the Black Sea, Bat et al. (2012) had been detected Pb and Cd levels lower than the recommended legal limits for human consumption according to the Turkish Food Codex (Anonymous, 2008). Chahid et al. (2014) determined the mean levels of Cd and Pb found in fish from Atlantic Sea (Morocco) as 0.009-0.036 mg/g for Cd and 0.013-0.014 mg/g for lead. The authors have concluded that these values fall within safe limits for human consumption.

Before the comparison, values were converted to mg/g wet weight. The mean muscle metal levels of *M. barbatus* were 0.553 mg/g for Cu, 0.0569 mg/g for Pb, and -0.0.014 mg/g for Cd. Mean values of Cu, Pb and Cd were below recommended limits of the Food and Agriculture Organization/World Health Organization (FAO/WHO, 2011): Cu: 30, Pb: 2 and Cd: 0.5 mg/g (ww). These values reached acceptable levels for hu-



**Fig 4: Differences of Cd concentrations in *Mullus barbatus* versus seasons.**

**Sl. 4: Razlike v koncentraciji Cd pri vrsti *Mullus barbatus* v različnih sezona.**

man consumption and with any health problems for consumers. Additionally, such values recorded in *M. barbatus* showed that the Syrian marine waters are not strongly polluted by anthropogenic activities. However, these activities are generally located along the coast and underwent also ship dismantling and heavy ship traffic which could affect the aquatic species living in the area. Some species, as *M. barbatus* display a high commercial value, and regularly and frequently measurements of heavy metals should be done in these species generally consumed as food to avoid also a negative impact on the local economy.

## VSEBNOST TEŽKIH KOVIN V TKIVIH BRADAČA, *MULLUS BARBATUS* (MULLIDAE) IZ SIRSKE OBALE (VZHODNO SREDOZEMSKO MORJE)

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### *POVZETEK*

Avtorji so v mišicah in jetrih primerkov bradača *Mullus barbatus* Linnaeus, 1758, ujetih ob sirske obale avgusta 2019 in maja 2020, merili vsebnost kadmija, svinca in bakra. Ugotovili so, da so se težke kovine kopičile na različnih nivojih in v različnih tkivih bradača, pri čemer je se je več težkih kovin nakopičilo v neužitnih delih bolj kot v užitnih mišicah. Najvišje povprečne vrednosti svinca ( $0,164 \pm 0,098 \text{ } \mu\text{g/g}$ ), in bakra ( $8,69 \pm 2,75 \text{ } \mu\text{g/g}$  mokre teže) so bile izmerjene v jetrih. Vsebnost Cd, Pb in Cu, izmerjena v užitnem mesu mišic, je bila nižja kot maksimalna dovoljena vrednost po kriterijih FAO/WHO za človeško uporabo. Redosled koncentracije težkih kovin v bradaču je bil Cu> Pb> Cd. Spremembe v koncentraciji težkih kovin v tkivih bradača so primerjali v različnih okoljih in različnih sezонаh.

**Ključne besede:** Sirija, težke kovine, *Mullus barbatus*, bioakumulacija, vzhodno Sredozemsko morje

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