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HEAVY METALS ACCUMULATION IN PROCESSED FISH PRODUCTS AND RISK ASSESSMENT ANALYSIS ON HUMAN HEALTH

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Abstract

The present study aimed to evaluate the concentration of heavy metals (aluminium - Al, beryllium - Be, cadmium - Cd, arsenic - As, nickel - Ni, chromium - Cr, copper - Cu, lead - Pb, zinc - Zn, and mercury - Hg respectively) in several processed fish foods (canned tuna, sardines, herring, salmon, and mackerel respectively) and assess the risks manifested on human health due to products consumption in Romania. In this context, the estimated daily intake (EDI) and target hazard quotient (THQ) was calculated based on the concentration of elements analysed by using the ICP-MS technique, the average fish products consumption in Romania and the average weight of Romanian consumers. The registered values for EDI and THQ were below 1, fact that indicates there is no risk on consumer's health. However, continuous screening of heavy metals contamination of food products is needed, due to the on-going risk for elements accumulation.

Key words: *fish products, heavy metals, risk assessment.*

INTRODUCTION

Animal protein is a major component of human diets, due to their high content in essential amino acids (Langyan et al., 2022). The availability of proteins for human consumption is necessary to ensure food security (Boyd et al., 2022). Today, almost 20% of the global animal-based protein destined for human consumption is ensured by fish meat and fish products derived from aquaculture or fisheries practices (Boyd et al., 2022).

Even more, fish meat and fish products are also a great source of long chain omega-3 polyunsaturated fatty acids such as eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA) (Leroy et al., 2023). At the same time, given the increased anthropogenic pressures exercised on aquatic ecosystems, fish meat and fish products have been observed to accumulate different contaminants which can cause toxicity if transferred in humans via consumption. Hazardous substances such as heavy metals can counteract the health benefits generated by fish consumption due to their lack of biodegradability and accumulation tendency (Djedjibegovic et al., 2020). Exposure and accumulation of heavy metals in the human body has been linked to a series of brain pathologies such as Parkinson's and Alzheimer's disease, Wilson disease, as well as multiple sclerosis (Mitra et al., 2022).

In this context, to protect the health of citizens, the European Union has established maximum permitted levels of heavy metals in fish meat and fish products through the regulation (EC) No 1881/2006. The regulation also calls for constant and continuous monitoring of heavy metals concentration levels in food products in order to detect and prevent possible threats posed to human health.

Therefore, the aim of the present study was to evaluate the concentration levels of aluminium (Al), beryllium (Be), cadmium (Cd), arsenic (As), nickel (Ni), chromium (Cr), copper (Cu), lead (Pb), zinc (Zn), and mercury (Hg) respectively, in several processed fish foods (canned tuna, sardines, herring, salmon, and mackerel respectively), commercialised in the Romanian market. Further on, a risk assessment analysis was conducted according to Kin order to identify if there are any potential health problems generated by the consumption of the aforementioned fish products.

MATERIALS AND METHODS

Canned fish products were purchased from the local stores in Galati city, Romania. Samples of processed fish products (*n=42*) were collected as it follows: canned tuna in brine, canned herring in vegetable oil, canned sardines in vegetable oil, canned tuna pate, canned salmon pate and canned mackerel pate (Figures 1 and 2).

Figure 1. Sample collection of canned tuna in brine

Figure 2. Sample collection of canned tuna pate

The samples were analysed within the Rexdan Research Infrastructure. Thus, each sample was digested with nitric acid and hydrogen peroxide and further analyzed by ICP-MS technique (Figure 3).

The final aqueous solution was analysed by inductively coupled plasma with mass spectrometry (ICP-MS) with the Perkin Elmer NexION 2000 equipment. The following heavy metals were quantified in saples: Be, Zn, Cu, Al, As, Cr, Hg, Cd, Pb, Ni, respectively.

Figure 3. Digested samples

Risk assessment analysis

The estimated daily intake (EDI) and the target hazard quotient (THQ) were calculated to highlight potential risks posed on human health by the consumption of processed fish products. EDI and THQ were computed according to Kowalska et al. (2020) as it follows:

$$EDI\ (mg/kg/day) = \frac{EF\ x\ ED\ x\ IR\ x\ C}{WAB\ x\ ATn}$$

where:
- EF = Exposure frequency (365 days/year);
- ED = Exposure duration – average lifetime in Romania (74.2 years);
- IR = Fish and seafood ingestion rate (kg person/day) - 17.26 g per person/day;
- C = metal concentration in fish muscle (mg/kg);
- WAB = average body weight (kg) - 78.65 kg for adults (85.1 kg for males and 72.2 kg for females;
- ATn = Average exposure time for non-carcinogen (365 days/year x ED).

$$THQ = \frac{EF\ x\ ED\ x\ IR\ x\ C}{RfD\ x\ WAB\ x\ ATn}\ x\ 10^{-3}$$

where:
- EF = Exposure frequency (365 days/year);
- ED = Exposure duration - average lifetime (74.2 years);
- IR = fish and seafood ingestion rate (g/day);
- C = metal concentration in edible tissue muscle (mg/kg);

- RfD = oral reference dose for each metal (mg/kg/day);
- WAB = average body weight (kg);
- ATn = average exposure time for non-carcinogen (365 days/year x ED).

RESULTS AND DISCUSSIONS

The analysis revealed that Be was below the detection limit in all analysed samples.

Further on, the following heavy metals accumulation trend in the canned tuna in brine was identified: Zn>Cu>Al>As>Cr>Hg>Cd>Pb>Ni (Figure 4). Zn had the highest concentration (2.798± 0.543 µg/g), while the lowest concentration was observed in case of Ni (0.018±0.002 µg/g

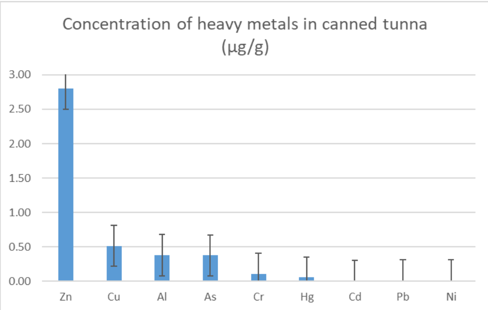

Figure. 4. Concentration of heavy metals in canned tuna in brine

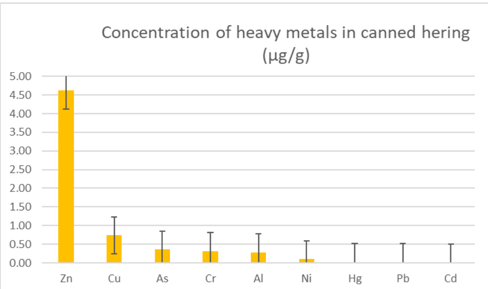

Figure. 5. Concentration of heavy metals in canned herring in vegetable oil

In case of heavy metals accumulation in canned herring in vegetable oil, the following trend was identified: Zn>Cu>As>Cr>Al>Ni>Hg>Pb>Cd (Figure 5). As previously observed the highest concentration level is in case of Zn (4.621±0.145 µg/g), while the lowest is observed in case of Cd concentration (0.002±0.001 µg/g).

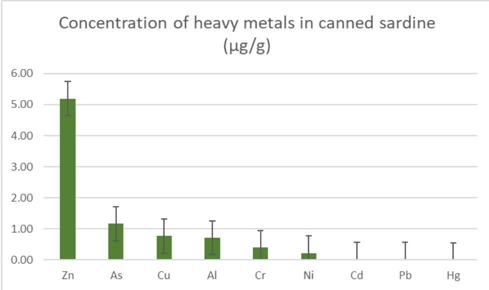

Figure. 6. Concentration of heavy metals in canned sardines in vegetable oil

In case of heavy metals accumulation in canned sardines in vegetable oil, the following accumulation trend was registered: Zn>As>Cu>Al>Cr>Ni>Cd>Pb>Hg. Zn had the highest concentration level (5.191±0.027 µg/g), while Hg registered the lowest concentration level (0.011±0.001 µg/g).

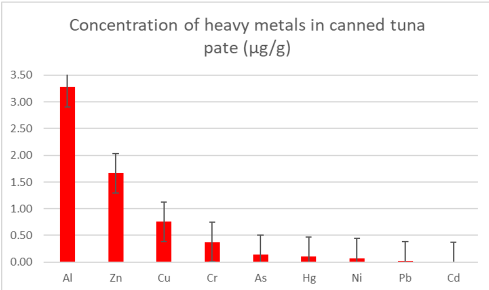

Figure. 7. Concentration of heavy metals in canned tuna pate

The accumulation trend of heavy metals in canned tuna pate is as it follows: Al>Zn>Cu>Cr>As>Hg>Ni>Pb>Cd (Figure 7). It can be observed that the highest concentration level was obtained in case of Al (3.275±0.205 µg/g). This can be due to poor quality of can material and transfer of Al from the packaging to the food inside. The lowest concentration level was registered in case of Cd (0.0042±0.0001 µg/g), as previously observed in canned herring.

Further on, in case of heavy metals accumulation in canned salmon pate, the following trend was identified: Al>Zn>Cu>Cr>As>Ni>Pb>Hg>Cd (Figure 8). As previously observed, the highest concentration level was in case of Al (3.803±0.159 µg/g), while the lowest concentration level was registered in case of Cd (0.0030±0.000 µg/g).

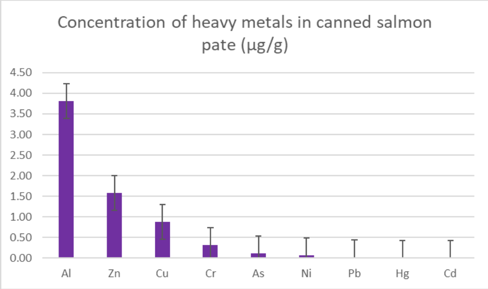

Figure. 8. Concentration of heavy metals in canned salmon pate

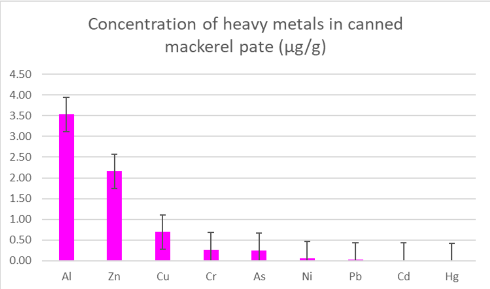

Figure. 9. Concentration of heavy metals in canned mackerel pate

The accumulation trend of heavy metals in canned mackerel pate was as it follows: Al>Zn>Cu>Cr>As>Ni>Pb>Cd>Hg (Figure 9). The highest value was registered in case of Al concentration (3.533±0.177 µg/g), while the lowest value was registered in case of Hg concentration (0.011±0.000 µg/g).

The results highlight that Zn had the tendency to accumulate in canned fish products conserved in vegetable oil or in brine. These results are in accordance with the study conducted by Islam et al., in which canned tuna, sardines and mackerel had the highest concentration levels in case of Zn, after magnesium (Islam & Mustafa, 2023). In all studied canned fish pates, the highest tendency of heavy metals accumulation was observed in case of Al.

Heavy metals with the lowest accumulation tendency were Cd, Hg and Ni.

As well, the concentrations of heavy metals in the canned fish samples registered values below the maximum permitted level set by the European Union in the (EC) No 1881/2006, which are 0.3 mg/kg in case of Pb, 0.05 mg/kg in case of Cd and 0.5 mg/kg in case of Hg.

In the study conducted by Miedico et al., the authors highlighted that 11% of the canned tuna samples exceeded the maximum permitted level in terms of Hg content and one tuna can exceed the maximum permitted levels in case of Cd content (0.22 mg/kg) (Miedico et al., 2020).

As well, El-Dahman et al. (2019) showed that canned tuna had higher Hg concentrations compared to other type of preserved fish (salted or frozen fish) (El-Dahman et al., 2019). In Table 1, the values of the computed estimated daily intake (EDI) are represented. As it can be observed, the estimated intake of Al from consuming canned fish pates is above the tolerable limits specified by World Health Organization, which can lead to chronic toxicity in the human body.

The same phenomenon was observed in case of Cd, which registered values of EDI above the tolerable limit, except for herring in vegetable oil and salmon pate. The intake of Ni was also above the tolerable limit in herring and sardines conserved in vegetable oil, and in tuna and salmon pates respectively. The intake of Hg is above the tolerable limit in all studied canned fish products. The most concerning heavy metal intake is registered in case of Pb, because the EDI values in all canned fish products were above the 0.0006 mg/kg/day, which is the intake that generated the loss in IQ by 1 point in children (Joint FAO/WHO, 2011).

The registered values for EDI are in accordance to the ones registered by (Djedjibegovic et al., 2020).

In Table 2 are represented the values of the computed target hazard quotient (THQ), which are significantly lower than 1. Scores of THQ lower than 1 indicate that there are no risks on the exposed population via the consumption of canned fish products. This is due to the low fish products consumption in Romania.

Table 1. Estimated daily intake (EDI) of heavy metals (mg/kg/day)

	Tuna in brine	Herring in oil	Sardines in oil	Tuna pate	Salmon pate	Mackerel pate	Tolerable limits
Al	0.0835	0.0624	0.1571	0.7189	0.8347	0.7754	0.28
As	0.0833	0.0782	0.2564	0.0302	0.0251	0.0548	-
Cd	0.0026	0.0005	0.0060	0.0009	0.0007	0.0035	0.00089
Cr	0.0241	0.0685	0.0886	0.0827	0.0706	0.0592	-
Cu	0.1128	0.1628	0.1695	0.1662	0.1935	0.1523	0.5
Ni	0.0040	0.0216	0.0496	0.0162	0.0163	0.0121	0.013
Pb	0.0040	0.0044	0.0056	0.0047	0.0049	0.0051	0.0006
Zn	0.6141	1.0140	1.1392	0.3655	0.3475	0.4738	1
Hg	0.0129	0.0046	0.0024	0.0224	0.0007	0.0024	0.00057

Table 2. Target hazard quotient (THQ)

	Tuna in brine	Herring in oil	Sardines in oil	Tuna pate	Salmon pate	Mackerel pate
Al	8E-08	6E-08	2E-07	7E-07	8E-07	8E-07
As	3E-04	3E-04	8E-04	1E-04	8E-05	2E-04
Cd	3E-06	5E-07	6E-06	9E-07	7E-07	3E-06
Cr	8E-06	2E-05	3E-05	3E-05	2E-05	2E-05
Cu	3E-06	4E-06	4E-06	4E-06	5E-06	4E-06
Ni	4E-07	2E-06	4E-06	1E-06	1E-06	1E-06
Pb	1E-06	1E-06	2E-06	1E-06	1E-06	1E-06
Zn	2E-06	3E-06	4E-06	1E-06	1E-06	2E-06
Hg	1E-04	5E-05	2E-05	2E-04	7E-06	2E-05

CONCLUSIONS

The main conclusion of the present study is that the concentrations of the studied heavy metals (Al, As, Cd, Cr, Cu, Ni, Pb, Zn, Hg) in canned fish products from Galati, Romania did not exceed the maximum permitted levels imposed by the European Union legislation.

Zn had the tendency to accumulate in canned fish products conserved in vegetable oil or in brine, while all studied canned fish pates, had the highest tendency to accumulate Al.

The estimated daily intake factor revealed that the consumption of canned fish products from Galati, Romania can contribute to the exposure of human population through diet to Al, Hg, Ni and Cd.

The target hazard quotient was significantly below 1, which indicates that there are no risks posed on the health of human consumers of canned fish products.

Continuous and constant monitoring of heavy metals in food products is recommended in order to ensure food safety and to protect the health of EU citizens.

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