



Risk Assessment of Total Mercury (T-Hg) in Commercial Seafood Marketed in Bangkok, Thailand

Irwan Ramadhan Ritonga¹, Sujaree Bureekul^{1,2,*}, Penjai Sompongchaiyakul^{1,2}

¹ Department of Marine Science, Faculty of Science, Chulalongkorn University, Bangkok, Thailand

² Center of Excellence on Hazardous Substance Management, Chulalongkorn University Research Building, Bangkok, Thailand

* Corresponding author: bsujaree@gmail.com

Article History

Submitted: 12 May 2021/ Revision received: 4 September 2021/ Accepted: 8 January 2022/ Published online: 29 March 2022

Abstract

Seafood is recognized as the high protein source of human consumption. However, it is hampered by mercury contamination. The objectives of this study are to determine total mercury (T-Hg) levels in edible portions of commercial seafood available in Bangkok's supermarkets, and to evaluate the potential risks from mercury through seafood consumption. Total 32 species, including 22 fish, 4 cephalopod and 6 shellfish, were purchased from supermarkets. Fish samples were dissected in 3 parts comprised of flesh, gill and viscera. While, cephalopod and shellfish were separated for edible tissues. The samples were digested in hot acid and were determined using cold-vapor atomic absorption spectrometry technique. The results revealed that T-Hg contained in the fish flesh > cephalopod > shellfish. In addition, T-Hg was accumulated in flesh > viscera > gill. Statistical analysis suggested that T-Hg accumulated in flesh was related positively with species, feeding habit, and habitat ($p < 0.05$). For risk analysis, estimated daily intake (EDI) of flesh ranged from 0.01 to 0.42 $\mu\text{g kg}^{-1}$ bodyweight d^{-1} , the lowest and highest EDI values were in salmon and yellowfin tuna, respectively. Yellowfin tuna, narrow-barred Spanish mackerel, fourfinger threadfin, and silver sillago were the 4 species that having the EDI values of T-Hg higher than the FAO/WHO recommended provisional tolerable daily intake (PTDI) of 0.23 $\mu\text{g kg}^{-1}$ bodyweight d^{-1} . Thus, the frequent consumption of these seafood are not recommended. In addition, the daily consumption of seafood should not exceed the maximum safe daily intake (MSDC). The MSDC of seafood in this study ranged from 15.5 (yellowfin tuna) to 474 (salmon) g d^{-1} .

Keywords: Flesh; Gill; Viscera; PTDI; MSDC

Introduction

Seafoods are a major source of nutrients, including protein, amino acids, vitamins, minerals

and omega-3 fatty acids, with significant roles in human health maintenance [1]. However, the presence of toxic chemicals in seafood tends to

discourage regular consumption. Thus, there is a need to communicate the benefits and risks of consuming these foods such as mercury (Hg), a lipophilic contaminant and therefore transferred in aquatic food webs through dietary fat.

The major Hg sources are either naturally occurring (volcanoes, geothermal activities, re-suspension from the earth crust) or anthropogenic activities, including agriculture, coal burning, mining, smelting, aluminium productions, cement manufacturing, dental amalgam, garbage disposal, oil refining, power plant and smelters [2]. These activities often tend to increase mercury pollution levels in water bodies, sediments and organisms in aquatic environments. Upon entering water column, Hg undergoes biogeochemical processes causing sinking to the seabed, conversion to methylmercury (methyl-Hg) by sulphate-reducing bacteria, and bioaccumulation and transfer into aquatic food chain [3].

Mercury accumulation attains the highest level in top predator fish tissues, through a process known as biomagnification [4]. Thus, fish consumption is one of the major routes for Hg exposure to human. Hg entering human body is easily absorbed into blood and transported to brain barrier through the methionine uptake system [5]. Due to the strong affinity for sulphhydryl groups in amino acid groups, it has the capacity to accumulate in both tissue and the central nervous system and therefore poses potential risks to human health, including heart disease, cardiovascular, central nervous system and fetus [6]. The adverse effects on human health for mercury were documented especially in the vulnerable groups such as pregnant women and children [7]. The hair samples from pregnant women analysed for total mercury were found positively associated with types of fish consumptions and was observed higher total mercury in pregnant women consumed canned fish [8]. In children who frequently consumed specific types of seafood such as sardine or mackerel

fish were reported to have higher mean blood mercury concentration [9]. The permissible limit for safe consumption of total Hg (T-Hg) contained in fish tissue is set at $0.5 \mu\text{g g}^{-1}$ wet weight for all fish and $1 \mu\text{g g}^{-1}$ wet weight for some predatory fish [10]. Therefore, everyone, especially pregnant women, nursing mothers and children, are advised to know specific information regarding fish and to avoid over seafood consumption [11].

Mercury in seafood has been reported across different Thailand coastal areas [12–13]. However, only certain organs in a few species of fish and other seafood have been surveyed. This study therefore investigates T-Hg in edible portions of commercial seafood from Bangkok markets and estimates the health risk of T-Hg contamination, through seafood consumption.

Materials and methods

1) Sample collection and preparation

Thirty-two seafood species from fish (22 species), cephalopods (4 species) and shellfish (6 species) bought in Bangkok supermarkets, Thailand, during February 2018. The common name, scientific name, food habit, and habitat of these organisms were then identified according to the fish identification book [14] and the website: <http://www.fishbase.org> [15] (Table 1). Subsequently, flesh, gill and viscera were separated from the fish samples, while only soft tissue was collected for cephalopods and shellfish. During the analysis, several samples were composited due to small sample size. All dissected samples were homogenized, placed in plastic bags and deep frozen, prior to freeze-drying in Lyopro 6000 instrument (HETO). The dried samples were then grounded into powder using an agate mortar and stored in a desiccator until chemical analysis. This was followed by calculating the moisture percentage of each individual sample based on weight, before and after drying.

Table 1 Scientific name, common name, habitat and food habit of seafood samples collected from Bangkok markets (n: the number of fish analyzed)

Scientific name	Common name	n	Habitat	Food habit
Fish				
<i>Alepes djedaba</i>	Shrimp scad	3	Pelagic	Carnivore
<i>Lactarius lactarius</i>	False trevally	1	Pelagic	Carnivore
<i>Lates calcarifer</i>	Barramundi	1	Pelagic	Carnivore
<i>Rastrelliger kanagurta</i>	Indian mackerel	3	Pelagic	Carnivore
<i>Salmo</i> spp.	Salmon	2	Pelagic	Carnivore
<i>Scomber japonicus</i>	Chub mackerel	3	Pelagic	Carnivore
<i>Scomberomorus cavalla</i>	King mackerel	1	Pelagic	Carnivore
<i>Scomberomorus commerson</i>	Narrow-barred Spanish mackerel	2	Pelagic	Carnivore
<i>Sphyraena putnamae</i>	Sawtooth barracuda	2	Pelagic	Carnivore
<i>Thunnus albacares</i>	Yellowfin tuna	1	Pelagic	Carnivore
<i>Thunnus tonggol</i>	Longtail tuna	1	Pelagic	Carnivore
<i>Eleutheronema tetradactylum</i>	Fourfinger threadfin	1	Pelagic	Omnivore
<i>Parastromateus niger</i>	Black pomfret	3	Pelagic	Omnivore
<i>Seriola quinqueradiata</i>	Japanese amberjack	3	Pelagic	Omnivore
<i>Epinephelus bleekeri</i>	Dusky tail Grouper	2	Demersal	Carnivore
<i>Epinephelus sexfasciatus</i>	Sixbar grouper	5	Demersal	Carnivore
<i>Larimichthys polyactis</i>	Small yellow croaker	1	Demersal	Carnivore
<i>Nemipterus nematophorus</i>	Double whip threadfin bream	6	Demersal	Carnivore
<i>Seriola nigrofasciata</i>	Black Banded trevally	2	Demersal	Carnivore
<i>Cynoglossus arel</i>	Largescale tonguesole	2	Demersal	Omnivore
<i>Mallotus villosus</i> *	Capelin	11	Demersal	Omnivore
<i>Sillago sihama</i> *	Silver Sillago	4	Demersal	Omnivore
Cephalopods				
<i>Architeuthis</i> spp.	Giant squid	2	Demersal	Carnivore
<i>Loligo duvauceli</i> *	Indian squid	3	Demersal	Carnivore
<i>Loliolus</i> spp.*	Kobi squid	10	Demersal	Carnivore
<i>Photololigo chinensis</i> *	Splendid squid	3	Demersal	Carnivore
Shellfish				
<i>Penaeus merguensis</i> *	White prawn	36	Demersal	Omnivore
<i>Penaeus monodon</i> *	Tiger shrimp	3	Demersal	Omnivore
<i>Meretrix meretrix</i> *	Asiatic hard clam	28	Benthic	Filter feeder
<i>Paphia textile</i> *	Textile venus	30	Benthic	Filter feeder
<i>Saccostrea</i> spp.*	Oyster	33	Benthic	Filter feeder
<i>Solen</i> spp.*	Razor clam	25	Benthic	Filter feeder

Remark: *The samples were composited before analyzing.

2) Reagents and laboratory wares

Milli-Q ultrapure water (>18.2 MΩ-cm) was prepared by Millipore Milli-Q lab water system and used in all preparation and digestion steps. All glassware and plastic bottles were pre-cleaned by soaking overnight in 10% (v/v) nitric acid

(HNO₃), rinsing thoroughly with milli-Q water and drying, prior to use. Supra-pure nitric acid and hydrochloric acid (HCl) were prepared by sub-boiling distillation from AR grade acids. Potassium bromide (KBr), potassium bromate (KBrO₃), hydroxylammonium chloride and other

chemicals using for sample preparation were AR grade chemicals.

Bromine chloride (BrCl) solution was prepared by adding 20 mL of Milli-Q water, 1.5 g KBrO₃, 1.1 g KBr, and 100 mL conc. supra-pure HCl in a pre-cleaned 125 mL Teflon bottle. Hydroxylamine solution was prepared by weighing and mixing 30 g of hydroxylammonium chloride and 70 g of Milli-Q water to yield a 30 % solution in a pre-cleaned 125 ml Teflon bottle. Also, sigma-Aldrich (Taufkirchen, Germany) standard solution of Hg (1000 mg L⁻¹) was used to prepare a working standard solution in the range of 0.3 to 10 µg L⁻¹ by serial dilution, in 3% HNO₃.

3) Sample preparation and digestion for mercury analyses

For the digestion, 0.200 g of dried samples were weighed accurately by analytical balance (Sartorius AX224) and placed into pre-cleaned test tubes. Subsequently, samples were digested by 1 mL of HNO₃ and 0.5 mL of H₂SO₄ in block heating system at 90–95°C, for 30 min. The digested solutions were then left to cool before dilution to 40 mL by adding 38 mL of 0.02N BrCl solution. This was followed by Hg analysis, using cold-vapor atomic absorption spectrometry (CV-AAS). Same procedure was repeated for reagent blanks and certified reference materials (CRMs).

The final digested solution then, introduced into Perkin Elmer® Flow Injection Mercury System (FIMS) model 400 with an AS-90 auto-sampler. 0.2% (w/v) NaBH₄ (Sodium borohydride) in 0.05% NaOH (sodium hydroxide) solutions were used as a reducing agents, while

the carrier solution was 3% (v/v) HCl. An argon stream was used as the carrier gas transporting Hg vapor into the absorption cell, while a hollow cathode mercury lamp operated at 253.7 nm for Hg determination. The working standard solutions of 0.3, 0.5, 1, 3, 5, 8, 10 µg-Hg L⁻¹ were also prepared fresh daily. A correlation coefficient (r) at 0.999 was compiled in order to continue operation for sample analysis. The T-Hg levels are expressed in µg g⁻¹ on wet weight basis, by converting dry weight to wet weight mass formula [16] as shown in Eq. 1.

4) Quality assurance and quality control (QA/QC)

For each batch, reagent blanks and CRMs were prepared and digested in parallel. The determine limit of detection (LOD) and limit of quantitation (LOQ) calculated by 3-times and 10-times of the average of standard deviation (SD) from 10 reagents blank, were at 0.003 µg g⁻¹ and 0.011 µg g⁻¹, respectively. The precision of an analytical procedure was complied with the analysis of an approximately 10% percent of all samples replication. The percentage of relative standard deviation (RSD) was reported within 12%. The accuracy of an analytical procedure was verified by CRMs including DORM-4 (dogfish muscle), TORT-3 (lobster hepatopancreas), and BCR-422 (Cod muscle). The recoveries of T-Hg were determined using Eq. 2.

The percentage of the CRMs recovery presented in Table 2, indicating a good agreement between certified and determined values. This verifies the accuracy and reliability of data obtained in this study.

$$\mu\text{g g}^{-1} \text{ wet weight} = \frac{(100 - \% \text{ moisture}) \times (\mu\text{g g}^{-1} \text{ dry weight})}{100} \quad (\text{Eq. 1})$$

$$\text{Recovery rate} = \frac{\text{Determined value}}{\text{Certified value}} \times 100 \quad (\text{Eq. 2})$$

Table 2 Results of T-Hg determination in CRMs

Certified Reference Materials	n	Certified value ($\mu\text{g g}^{-1}$)	Determined value ($\mu\text{g g}^{-1}$)	Recovery rate
DORM-4 (Dog fish)	6	0.412±0.036	0.377±0.017	91.5
TORT-3 (Lobster Hepatopancreas)	4	0.292±0.022	0.278±0.009	95.2
BCR-422 (Cod muscle)	5	0.559±0.016	0.443±0.021	79.2

5) Health risk assessment

It is well recognized that fish consumption is the most important pathway for mercury exposure to human [2, 5]. Mercury had the adverse effect to human health particularly response to the heart disease, cardiovascular, central nervous system and fetus effect [6]. In fish flesh, mercury contamination is recommended to not exceed the $0.5 \mu\text{g g}^{-1}$ with the exception of predator fish that could be contained up to $1.0 \mu\text{g g}^{-1}$ [10]. In addition, the mercury exposure via daily human ingestion should not be greater than provisional tolerable daily intake (PTDI) or the toxicological reference value. To evaluate risk of seafood consumption for adult's consumer in Thailand, the estimated daily intake (EDI) values of Hg exposure from the flesh of each samples were calculated based on Eq. 3 [7].

$$EDI = \frac{C_m \times FIR \times EF \times ED}{BW \times AT} \quad (\text{Eq. 3})$$

Where, methyl-Hg levels (C_m) in determined seafood is accounted for 93% of T-Hg in fish flesh, using the value from the Windom and Cranmer [13] and Anual et al., [16]. Daily Fish Ingestion Rate (FIR) of seafood is 28.7 g meal^{-1} [17]. Exposure frequency (EF) is assumed for the fish consumption of 1 meal per day which is equivalent to 365 meals per year. Exposure duration (ED) is 72.05 years, the average lifetime of adults in Thailand) [18]. The average body weight (BW) of adults in Thailand is 56 kg [19]. The average exposed time (AT) is 72.05 years for non-carcinogenic substance and is multiplied by 365 days. As the consequence,

an amount for safety of daily seafood consumption by adult's consumers in Thailand can be calculated using Eq. 4 [7] for the maximum safe daily consumption (MSDC) in g d^{-1} .

$$MSDC = \frac{PTDI \times BW}{C_m} \quad (\text{Eq. 4})$$

Where, PTDI was applied as toxicological reference value at $0.23 \mu\text{g kg}^{-1} \text{ body weight d}^{-1}$ [20].

6) Statistical analysis

The descriptive statistics including minimum, maximum, average and standard deviation were calculated and reported for T-Hg in analysed seafood. The number of species studied in this work is 32, therefore, the Shapiro-Wilk has been chosen to test the distribution's normality. Meanwhile, Levene's test was applied to verify the data's homogeneity (SPSS version 19.0). Due the data set was not homogenous and did not have a normal distribution, a non-parametric ANOVA (analysis of variance), Kruskal-Wallis test (one-way ANOVA on ranks) and Mann Whitney were selected to test for any significant difference between groups. A statistically significant difference was designated in cases where $p < 0.05$.

Results and discussion

1) Total mercury (T-Hg) levels in fish flesh

The range of T-Hg levels in flesh from 22 marketed fish species in Bangkok were 0.02 to $0.89 \mu\text{g g}^{-1}$ wet weight, with a $0.29 \pm 0.25 \mu\text{g g}^{-1}$ average (Figure 1 and Supplementary Material

(SM) 1). Yellowfin tuna contained the highest T-Hg levels of $0.89 \mu\text{g g}^{-1}$ wet weight while salmon flesh had the lowest values of $0.03 \mu\text{g g}^{-1}$ wet weight. According to the standard guidelines of European and Thailand Commission Regulations [10, 21], the permissible T-Hg level in fish flesh is anything below $0.5 \mu\text{g g}^{-1}$ of wet weight. It was found that yellowfin tuna, narrow-barred Spanish mackerel, four finger threadfin, and silver sillago were the 4- species with T-Hg levels above the recommended values. However, the allowance for the predatory for fish like the yellowfin tuna was set to not be higher than $1.0 \mu\text{g g}^{-1}$ wet weight. Therefore, T-Hg levels in marketed yellowfin tuna were presented within the recommended values (Figure 1).

The yellowfin tuna T-Hg levels reported in this study were 29 times higher, compared to salmon. This findings are in accordance with a separate study from Slovenian market, reporting a 15 times higher value in yellowfin tuna, compared to salmon obtained from the same

market [22]. In addition, the T-Hg levels in yellowfin tuna in this study ($0.89 \mu\text{g g}^{-1}$ wet weight) was higher than observed from other places around the globe at the range between 0.04 to $0.60 \mu\text{g g}^{-1}$ wet weight in flesh [23]. These two fishes are both pelagic carnivore, however, the highest T-Hg levels in yellowfin tuna are possibly related to the fish's predatory life cycle in the food webs, fast swimming and relatively high metabolism, bound to encourage Hg accumulation and magnification in their body [23]. Meanwhile, salmon's feeding habit and habitat allow exposure to low mercury contamination. A study reported that the cultivated salmon had lower mercury content, compared to wild salmon [24]. This is possibly due to growth dilution in rapidly growing cultivated fish, as well as a dilution of Hg with high lipid content in cultivated fish [25]. However, a controversial study reported farmed salmon to contain more Hg, compared to wild counterparts [26].

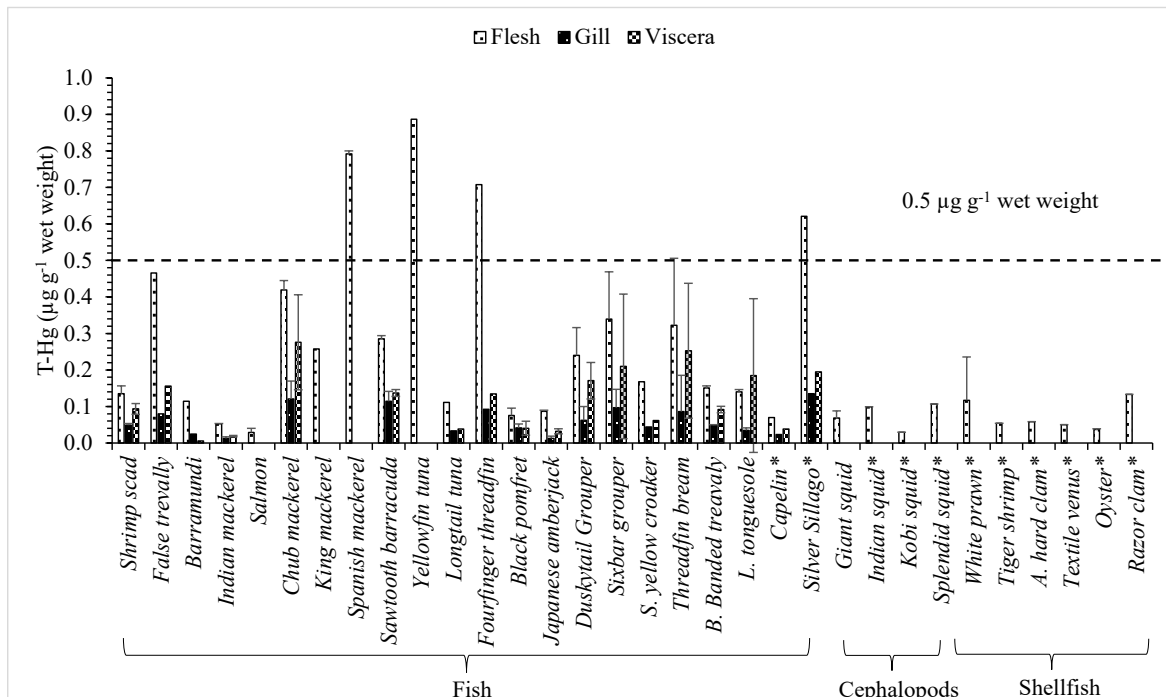


Figure 1 Average and standard deviation of T-Hg ($\mu\text{g g}^{-1}$ wet weight) levels in each species of seafood in Bangkok supermarkets comparing to the recommended maximum mercury level (dash lines) for fish and fish products by the Ministry of Public Health of Thailand (MPH, 2020) and the European Commission Regulation (EC, 2006). Star symbols (*) represents composite samples before analyzing.

2) Comparison of total mercury (T-Hg) levels in flesh, gill and viscera of fish

Concentration of T-Hg levels in gill and viscera from 22 fish species marketed in Bangkok ranged between 0.01–0.13 and 0.01–0.28 $\mu\text{g g}^{-1}$ wet weight, with an average of 0.06 ± 0.04 and 0.12 ± 0.08 $\mu\text{g g}^{-1}$ wet weight, respectively (Figure 1, SM 1). Generally, the T-Hg accumulation pattern among different fish tissues was found to be in the order from flesh > viscera > gill, with a mean concentration (SM 1). The results showed flesh tissue contained about twice and quadruple the content in viscera and gill. According to the Kruskal Wallis test, the T-Hg levels were significantly different in fish tissue (Chi-Square = 41.3, $p < 0.05$), (SM 2a).

A study reported that fish flesh has the highest T-Hg levels probably due to the strong complexation with thiol, a predominant organosulfur compound and likely part of a larger peptide (for instance, glutathione) or protein [27]. Meanwhile, gills was observed as the first targeted organ to react with mercury suspended from surrounding water column and sediment, through ion transport, gas exchange, acid-base regulation and waste excretion [28]. Upon entering through gill adsorption and food digestion, mercury is absorbed and accumulated in tissues, through blood circulation, and subsequently, tightly bound to amino groups, nitrogen, as well as sulphur, to form stable complexes in fish tissues, and stored, particularly in flesh [29]. The high levels of T-Hg observed in flesh, compared to other tissues is possibly influenced by the presence of metallothionein's protection against oxidative stress and neuroprotective mechanism. This detoxification allowed the homeostatic regulation of metals [30] that may accumulate in tissue. This indicated the flesh's metallothionein levels are probably higher, compared to viscera and gill. Therefore, without detoxification mechanisms, flesh T-Hg are possibly stored longer and accumulate more, compared to other tissues.

Similar to the study from the Northern Gulf of California [31], this study observed the highest T-Hg levels in fish flesh, where total mercury content (0.44 ± 0.06 $\mu\text{g g}^{-1}$ wet weight) tended to be higher, compared to the gill (0.29 ± 0.04 $\mu\text{g g}^{-1}$), kidney (0.20 ± 0.07 $\mu\text{g g}^{-1}$), and liver (0.02 ± 0.004 $\mu\text{g g}^{-1}$). However, a study conducted in New Jersey, USA [32] reported higher T-Hg levels in kidney tissues (as part of viscera, 0.57 ± 0.09 $\mu\text{g g}^{-1}$ wet weight), compared to flesh (0.32 ± 0.02 $\mu\text{g g}^{-1}$), brain (0.09 ± 0.01 $\mu\text{g g}^{-1}$) and skin (0.32 ± 0.02 $\mu\text{g g}^{-1}$) tissues. This difference in trends is probably not only dependent on fish species, feeding habits and habitat but also on geographic location, age, size and Hg background in the environs [16, 31–32].

3) Total mercury (T-Hg) in cephalopods and shellfish

The range of T-Hg concentration in 4 cephalopod and 6 shellfish species were 0.03–0.11 and 0.04–0.13 $\mu\text{g g}^{-1}$ wet weight, with an average of 0.08 ± 0.04 and 0.07 ± 0.04 $\mu\text{g g}^{-1}$ wet weight, respectively (SM 1). Figure 1 displayed that all cephalopod and shellfish samples have T-Hg much lower than 0.5 $\mu\text{g g}^{-1}$ wet weight, and fish counterparts. Based on the Kruskal Wallis test (SM 2b), statistical analysis also suggested significant differences in correlation between T-Hg accumulation with fish flesh, cephalopods, and shellfish (Chi-Square = 13.8, $p < 0.05$). However, there was not a significantly difference in the results of cephalopods and shellfish ($p > 0.05$), from the Mann Whitney test (SM 3). In cephalopod and shellfish, the lowest T-Hg value was detected in kobi squid (0.03 $\mu\text{g g}^{-1}$ wet weight), while the highest was in razor clam (0.13 $\mu\text{g g}^{-1}$ wet weight). The high T-Hg contained in shellfish is probably related to its behavior; feeding habit and habitat. Clams use muscles for self-buried and to filter suspended particulates, including mercury, from both water column and sediments, through gills [33].

In this study, the T-Hg levels in flesh tissue within fish, cephalopods and shellfish were compared to some other regions (Table 3). Data from the literature showed that T-Hg levels varied widely in seafood, depending on sampling areas and species. Importantly, T-Hg levels in fish in this study were relatively higher, compared to other studies [16, 34–39]. While the shellfish and cephalopod T-Hg level were not higher, compared to other literatures [16, 34, 36, 40].

4) Total mercury (T-Hg) levels across food habit and habitat

The tendency for higher T-Hg accumulation in fish flesh, followed by shellfish and cephalopods was observed (SM 5), when all seafood species were categorized according to its feeding habit and habitat. For fish, the accumulation of T-Hg tended to decrease followed from carni-

vore demersal>carnivore pelagic>omnivores demersal>omnivore pelagic, respectively. Meanwhile, the trend in the cephalopods and shellfish was in the order, omnivore demersal>carnivore demersal>filter feeder benthic (SM 5). In general, carnivore demersal fish reported a 4 times higher T-Hg level in flesh compared to cephalopod and a 3 times higher, compared to filter feeder benthic. This finding is consistent with a study from Taiwan market, where carnivore fish ($0.07 \mu\text{g g}^{-1}$ wet weight) contained as twice higher T-Hg level, compared to non-carnivore fish ($0.03 \mu\text{g g}^{-1}$ wet weight). This difference of the T-Hg accumulation pattern was supported by the Kruskal-Wallis test (SM 4), where seafood samples collected from Bangkok markets were found to have a significant difference with food habit and habitat (Chi-Square = 15.5, $p < 0.05$).

Table 3 A comparison of T-Hg levels ($\mu\text{g g}^{-1}$ wet weight) in seafood collected from Bangkok markets and other areas (markets and wild caught)

Sample Area	Species	n	T-Hg ($\mu\text{g g}^{-1}$ wet weight)		References
			Mean	Range	
Huelva, Spain (Market)	Fish	8	0.23 ± 0.16	-	[34]
	Cephalopods	1	0.83	-	
	Shellfish	3	0.86 ± 0.88	-	
Taiwan (Market)	Fish	45	0.07	-	[35]
	Cephalopods and shellfish	51	0.03	-	
Cluj-Napoca, Romania (Market)	Fish	18	0.21 ± 0.17	-	[36]
	Cephalopods	9	0.11 ± 0.04	-	
	Shellfish	9	0.09 ± 0.06	-	
Map Ta Phut Industrial Estate, Thailand (Caught)	Fish	220	-	<0.004–0.19	[37]
	Shellfish	48	-	<0.004–0.09	
Italy (Market)	Fish	330	-	0.08–0.47	[38]
	Cephalopods	30	-	0.07	
	Shellfish	90	-	0.06	
Sanmen Bay, China (Caught)	Fish	33	-	0.00–0.02	[39]
	Shellfish	41	-	0.00–0.09	
West Peninsular, Malaysia (Market)	Fish	74	-	0.02–0.61	[16]
	Shellfish	15	-	0.01–0.12	
	Cephalopods	12	-	0.02–0.22	
Sonora, Mexico (Caught)	Fish	194	0.15 ± 0.19	<0.004–1.22	[40]
	Shellfish	44	0.16 ± 0.22	<0.004–1.12	
Bangkok, Thailand (Market)	Fish-Flesh	47	0.29 ± 0.25	0.02–0.89	This study
	Cephalopods	5	0.08 ± 0.03	0.03–0.11	
	Shellfish	8	0.08 ± 0.04	0.04–0.13	

The high T-Hg accumulated in carnivore demersal fish and cephalopods in this study is possibly influenced by the food habit and habitat in the food chain. A study investigated that demersal fish are more likely exposed to Hg, compared to pelagic fish, due to the close association with a relatively higher T-Hg level in sediments [41]. Additionally, fish species, trophic level, habitat, body size and age [4, 42] are also possibly influenced high levels in carnivore demersal fish within this study. Interestingly, the higher T-Hg levels observed in carnivore and non-carnivore fishes is also similar to report from other regions. A study conducted in Guanabara Bay, south-eastern Brazil reported the top predator fish ($0.77 \mu\text{g g}^{-1}$ dry weight) had the highest flesh mercury levels, followed by less voracious species ($0.43 \mu\text{g g}^{-1}$ dry weight) and planktivorous fish ($0.04 \mu\text{g g}^{-1}$ dry weight) [43]. Furthermore, carnivore fishes have the tendency to accumulate higher T-Hg levels, compared to non-carnivore counterparts, due to bioaccumulation and biomagnification of Hg content, from the food web [16, 42].

5) Exposure assessment for human consumers

Fish consumption is a choice to consider from one individual to another. The elemental intake from seafood ingestion is dependent on the concentration, and the amount consumed. The accumulation of T-Hg may have a direct effect on human health after daily consumption. Thus, it is ought to investigate the exposure assessment for seafood consumption.

Based on the risk assessment for human consumers, the EDI of mercury and MSDC for seafood flesh were calculated (Figure 2, SM 1). The EDI value in Bangkok seafood species ranged from 0.01 to $0.42 \mu\text{g kg}^{-1}$ body-weight d^{-1} , with a $0.11 \pm 0.11 \mu\text{g kg}^{-1}$ bodyweight d^{-1} average. The

average EDI values in fish species ($0.14 \pm 0.12 \mu\text{g kg}^{-1}$ bodyweight d^{-1}) was higher, compared to shellfish ($0.04 \pm 0.02 \mu\text{g kg}^{-1}$ bodyweight d^{-1}) and cephalopods ($0.04 \pm 0.02 \mu\text{g kg}^{-1}$ bodyweight d^{-1}). Figure 2 displayed the EDI of each species compared with the PTDI value at $0.23 \mu\text{g kg}^{-1}$ bodyweight d^{-1} [20]. There are 4 species namely yellowfin tuna, narrow-barred Spanish mackerel, fourfinger threadfin, and silver sillago having EDI values exceeding this recommendation. This indicates that frequently consumed some seafood species from Bangkok markets may potentially pose a health risk through consumption. Therefore, consumers ought to reduce mercury intake, by selecting less Hg-contained seafood, to avoid over-consumption beyond the PTDI values.

Conclusion

This study reported the T-Hg levels in commercial seafood (fish, cephalopods, and shellfish) with different fish tissues (flesh, gill and viscera) from Bangkok markets, Thailand. T-Hg levels ranged from 0.02–0.89, 0.03–0.11 and $0.04\text{--}0.13 \mu\text{g g}^{-1}$ wet weight, for fish flesh, cephalopods and shellfishes, respectively. In fish tissues, the highest T-Hg level was found in flesh, followed by viscera and gill. Furthermore, carnivore demersal fish were most likely to accumulate T-Hg, compared to other species. The T-Hg levels were also positively significantly correlated with fish tissues, seafood species, food habit and habitat. Therefore, restricted consumption of seafood with less than the PTDI value at $0.23 \mu\text{g kg}^{-1}$ bodyweight d^{-1} is recommended. The MSDC for yellowfin tuna, narrow-barred Spanish mackerel, fourfinger threadfin, and silver sillago are suggested to be below 15.5 g d^{-1} for adult consumption, to minimize the health risks.

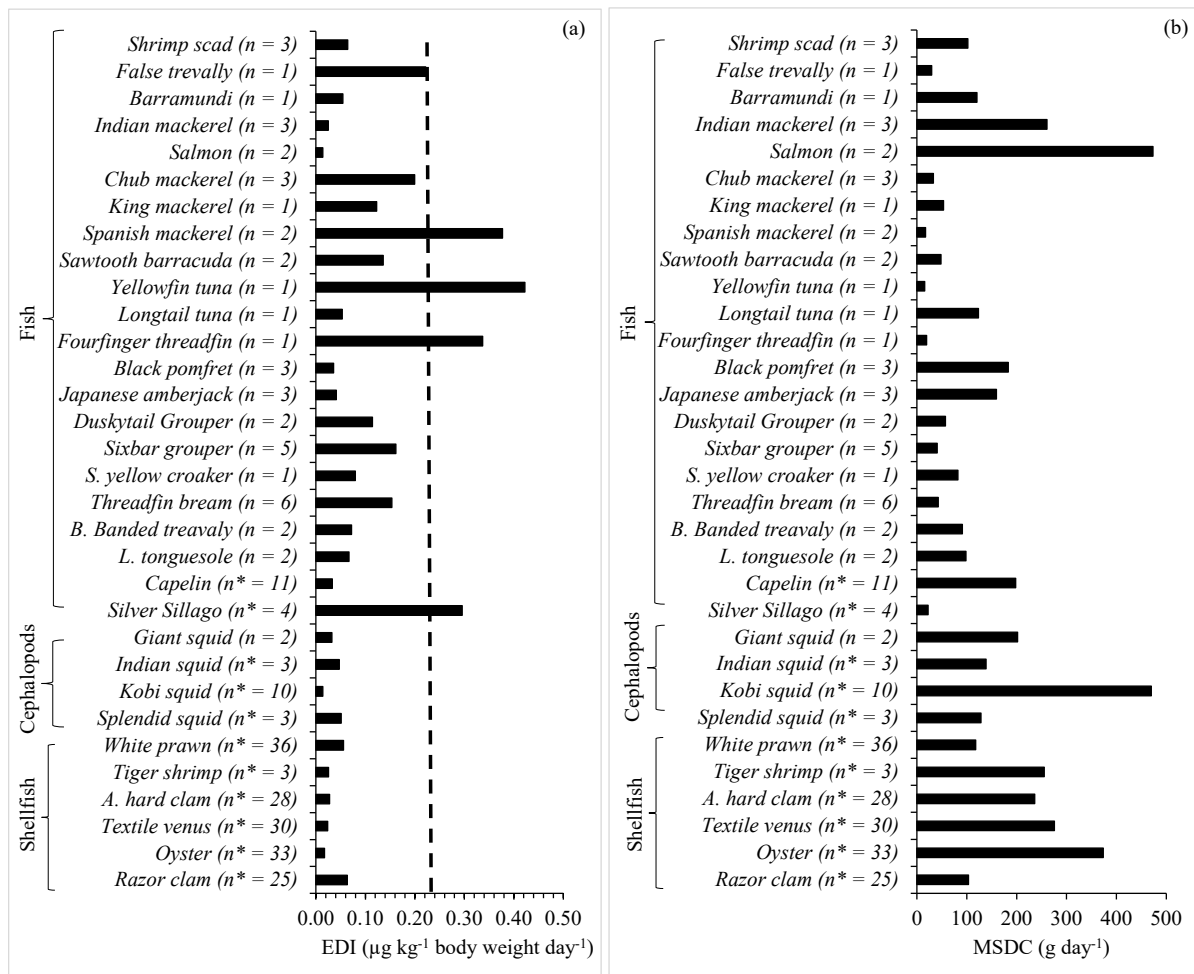


Figure 2 a) The values of EDI in $\mu\text{g kg}^{-1}$ body weight d^{-1} , b) MSDC in g d^{-1} of commercial seafood marketed from Bangkok markets. Vertical dash line represents the PTDI value at $0.23 \mu\text{g kg}^{-1}$ bodyweight d^{-1} for adult consumption. Star symbols (*) represents composite samples before analyzing.

Conflict of Interest: The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This study was financial supported by the Chulalongkorn University Graduate School Thesis Grant Batch 1/2020, and the Center of Excellence on Hazardous Substance Management (HSM), Contract No. HSM-PJ-CT-17-01. Chulalongkorn University. The authors also would like to thank P. Saensith, T. Ubonyaem, J. Kayee, and the members of the chemical

oceanography laboratory in Department of Marine Science, Faculty of Science, Chulalongkorn University.

References

[1] Hicks, C.C., Cohen, P.J., Graham, N. A.J.J., Nash, K.L., Allison, E.H., D’Lima, C., ..., MacNeil, M.A. Harnessing global fisheries to tackle micronutrient deficiencies. *Nature*, 2019, 574(7776), 95–98.

[2] United Nations Environment Programme (UNEP). Global mercury assessment 2013: Source, emissions, releases and environmental transport, UNEP Chemicals Branch, Geneva, Switzerland, 2013. [Online] Available

- from: <http://www.unep.org/hazardous-substances/Mercury/Informationmaterials/ReportsandPublications/tabid/3593/Default.aspx>. [Accessed 7 Mei 2021].
- [3] Parks, J.M., Johs, A., Podar, M., Bridou, R., Hurt, R.A., Smith, S.D., ..., Brandt, C.C. The genetic basis for bacterial mercury methylation. *Science*, 2013, 339, 1332–1335.
- [4] Liu, J., Cao, L., Dou, S. Trophic transfer, biomagnification and risk assessments of four common heavy metals in the food web of Laizhou Bay, the Bohai Sea. *Science of the Total Environment*, 2019, 670, 508–522.
- [5] Clarkson, T.W., Magos, L. The toxicology of mercury and its chemical compounds. *304 Critical Reviews in Toxicology*, 2006, 36(8), 609–662.
- [6] Kim, K.H., Kabir, E., Jahan, S.A. A review on the distribution of Hg in the environment and its human health impacts. *Journal of Hazardous Materials*, 2016, 310, 278–279.
- [7] United States Environmental Protection Agency (USEPA). Guidance for assessing chemical contaminant data for use in fish advisories. 3rd Edition. Washington: Office of Science and Technology Office of Water. 2000. [Online] Available from: <https://www.epa.gov/sites/production/files/2018-11/documents/guidance-assess-chemicalcontaminant-vol2-third-edition.pdf> [Accessed 9 Mei 2021].
- [8] Xue, F., Holzman, C., Rahbar, M.H., Trosko, K., Fischer, L. Maternal fish consumption, mercury levels, and risk of preterm delivery. *Environmental Health Perspectives*, 2007, 115(1), 42–47.
- [9] Rahbar, M.H., Samms-Vaughan, M., Loveland, K.A., Ardjomand-Hessabi, M., Chen, Z., Bressler, J., Shakespeare-Pellington, S., Grove, M.L., Bloom, K., Pearson, D.A., Lalor, G.C., Boerwinkle, E. Seafood consumption and blood mercury concentrations in Jamaican children with and without autism spectrum disorders. *Neurotoxicity Research*, 2013, 23 (1), 22–38.
- [10] European Commission Regulation (EC). Commission regulation no 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs. In *Official Journal of the European Union L364*. 2006. [Online] Available from: <https://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2006:364:0005:0024:EN:PDF> [Accessed 8 Mei 2021].
- [11] Environmental Protection Agency/United States Government (EPA). EPA-FDA Advice about eating fish and shellfish, USA: Environmental Protection Agency. 2017. [Online] Available from: <https://www.epa.gov/fish-tech/epa-guidance-developing-fish-advisories>. [Accessed 9 Mei 2021].
- [12] Thongra-Ar, W., Parkpian, P. Total mercury concentrations in coastal areas of Thailand: A review. *ScienceAsia*, 2002, 28(1), 301–312.
- [13] Windom, H.L., Cranmer, G. Lack of observed impacts of gas production of Bongkot Field, Thailand on marine biota. *Marine Pollution Bulletin*, 1998, 36(10), 799–807.
- [14] Ahmad, A., Lim, A., Khiok, P., Nor Azman, Z., Mohd Saki, N., Tassapon, K. Marine fishes and crustaceans of the Southeast Asian region. First edition. Terengganu, Malaysia: SEAFDEC/MFRDMD/SP/40, 2018, 1–238.
- [15] Froese, R., Pauly, D. World wide web electronic publication, 2019. [Online] Available from: www.fishbase.org [Accessed 9 Mei 2021].
- [15] Reeb, J., Milota, M. Moisture content by the oven-dry method for industrial testing. The Western Dry Kiln Association Meeting, Portland, USA: Corvallis, Or. : Western Dry

- Kiln Association, 1999, 9. [Online] Available from: https://ir.library.oregonstate.edu/concern/conference_proceedings_or_journals/fq977v732982 [Accessed 9 Mei 2021].
- [16] Anual, Z.F., Maher, W., Krikowa, F., Hakim, L., Ahmad, N.I., Foster, S. Mercury and risk assessment from consumption of crustaceans, cephalopods and fish from West Peninsular Malaysia. *Microchemical Journal*, 2018, 140, 214–221.
- [17] Needham, S., Funge-Smith, S.J. The consumption of fish and fish products in the Asia-Pacific region based on household surveys. Bangkok: RAP publication. 2015. [Online] Available from: <http://www.fao.org/3/a-i5151e.pdf> [Accessed 8 Mei 2021].
- [18] Porapakham, Y., Rao, C., Pattaraarchchai, J., Polprasert, W., Vos, T., Adair, T., Lopez, A.D. Estimated causes of death in Thailand, 2005: Implications for health policy. *Population Health Metrics*, 2010, 8(1), 1–11.
- [19] Mathuramon, P., Chirachariyavej, T., Peonim, A.V, Rochanawutanon, M. Correlation of internal organ weight with body weight and length in normal Thai adults. *Journal of the Medical Association of Thailand*, 2009, 92(2), 250–258.
- [20] The Joint FAO/WHO Expert Committee on Food Additives (JEFCA). Evaluation of certain food additives and contaminants: Sixty-seventh report of the Joint FAO/WHO Expert Committee on Food Additives. Rome, Italy: World Health Organization. 2007. [Online] Available from: https://apps.who.int/iris/bitstream/handle/10665/43645/9789241660587_eng.pdf [Accessed 8 Mei 2021].
- [21] Ministry of Public Health (MPH). Notification of Ministry of Public Health (No 414) B.E. 2563. Issued by virtue of Food Act B.E. 2522. Re: Standards for Contaminants in Food (Vol. 137). Thailand: Minister of Public Health. 2020. [Online] Available from: http://food.fda.moph.go.th/law/data/announ_moph/V.English/P414_E.pdf [Accessed 8 Mei 2021].
- [22] Miklavcic, A., Stibilj, V., Heath, E., Polak, T., Tratnik, J.S., Klavž, J., ..., Horvat, M. Mercury, selenium, PCBs and fatty acids in fresh and canned fish available on the Slovenian market. *Food Chemistry*, 2011, 124(3), 711–720.
- [23] Nicklisch, S.C.T., Bonito, L.T., Sandin, S., Hamdoun, A. Mercury levels of yellowfin tuna (*Thunnus albacares*) are associated with capture location. *Environmental Pollution*, 2017, 229, 87–93.
- [24] Lundebye, A. Lower levels of persistent organic pollutants, metals and the marine omega 3-fatty acid DHA in farmed compared to wild Atlantic salmon (*Salmo salar*). *Environmental Research*, 2017, 155, 49–59.
- [25] Jardine, L.B., Burt, M.D.B., Arp, P.A., Diamond, A.W. Mercury comparisons between farmed and wild Atlantic salmon (*Salmo salar* L.) and Atlantic cod (*Gadus morhua* L.). *Aquaculture Research*, 2009, 40(10), 1148–1159.
- [26] Foran, J.A., Good, D.H., Carpenter, D.O., Hamilton, M.C., Knuth, B.A., Schwager, S.J. Quantitative analysis of the benefits and risks of consuming farmed and wild salmon. *Journal of Nutrition*, 2005, 135 (11), 2639–2643.
- [27] Ulrich, K., Ursula, J. The role of thiols in antioxidant systems. *Free Radical Biology & Medicine*, 2016, 176(10), 139–148.
- [28] Evans, D.H., Piermarini, P.M., Choe, K.P. The multifunctional fish gill: Dominant site of gas exchange, osmoregulation, acid-base regulation, and excretion of nitrogenous waste. *Physiological Reviews*, 2005, 85(1), 97–177.
- [29] Harris, H.H., Pickering, I.J., George, G.N. The chemical form of mercury in fish. *Science*, 2003, 301(5637), 1203.

- [30] Wang, W.C., Mao, H., Ma, D.D., Yang, W.X. Characteristics, functions, and applications of metallothionein in aquatic vertebrates. *Frontiers in Marine Science*, 2014, 1, 1–12.
- [31] Acosta-Lizárraga, L.G., Bergés-Tiznado, M.E., Bojórquez-Sánchez, C., Osuna-Martínez, C.C., Páez-Osuna, F. Bioaccumulation of mercury and selenium in tissues of the mesopelagic fish Pacific hake (*Merluccius productus*) from the northern Gulf of California and the risk assessment on human health. *Chemosphere*, 2020, 255, 126941.
- [32] Burger, J., Jeitner, C., Donio, M., Pittfield, T., Gochfeld, M. Mercury and selenium levels, and selenium:mercury molar ratios of brain, muscle and other tissues in bluefish (*Pomatomus saltatrix*) from New Jersey, USA. *Science of the Total Environment*, 2013, 443, 278–286.
- [33] Amisah, S., Obirikorang, K.A., Adjei Boateng, D. Bioaccumulation of heavy metals in the volta clam, *Galatea Paradoxa* (Born, 1778) in relation to their geoaccumulation in benthic sediments of the Volta estuary, Ghana. *Water Quality, Exposure and Health*, 2011, 2(3–4), 147–156.
- [34] Bordajandi, L.R., Gómez, G., Abad, E., Rivera, J., Del Mar Fernandez-Baston, M., Blasco, ..., González, M.J. Survey of persistent organochlorine contaminants (PCBs, PCDD/Fs, and PAHs), heavy metals (Cu, Cd, Zn, Pb, and Hg), and arsenic in food samples from Huelva (Spain): Levels and health implications. *Journal of Agricultural and Food Chemistry*, 2004, 52(4), 992–1001.
- [35] Chen, Y.C., Chen, M.H. Mercury levels of seafood commonly consumed in Taiwan. *Journal of Food and Drug Analysis*, 2006, 14(4), 373–378.
- [36] Miclean, M., Cadar, O., Tanaselia, C., Gog, A., Senila, M., Groza, I.S. Levels of metals and organochlorine compounds in seafood consumed in Cluj-Napoca, Romania. *Environmental Engineering and Management Journal*, 2012, 11(1), 133–140.
- [37] Thongra-ar, W., Musika, C., Wongsudawan, W., Munhapon, A. Health risk assessment of heavy metals via seafood consumption from coastal area of Map Ta Phut Industrial Estate, Rayong Province, Burapha Science Journal, 2014, 19, 39–54.
- [38] Zaza, S., de Balogh, K., Palmery, M., Pastorelli, A.A., Stacchini, P. Human exposure in Italy to lead, cadmium and mercury through fish and seafood product consumption from Eastern Central Atlantic Fishing Area. *Journal of Food Composition and Analysis*, 2015, 40, 148–153.
- [39] Liu, Liao, Y., Shou, L. Concentration and potential health risk of heavy metals in seafoods collected from Sanmen Bay and its adjacent areas, China. *Marine Pollution Bulletin*, 2018, 131, 356–364.
- [40] Garcia-Hernandez, J., Ortega-Velez, M.I., Contreras-Paniagua, A.D., Aguilera-Marquez, D., Leyva-Garcia, G., Torre, J. Mercury concentrations in seafood and the associated risk in women with high fish consumption from coastal villages of Sonora, Mexico. *Food Chemistry Toxicology*, 2018, 120, 367–377.
- [41] Hatje, V., Andrade, R.L.B., Jesus, R.M., Masqué, P., Albergaria-Barbosa, A.C.R., de Andrade, J.B., Santos, A.C.S.S. Historical records of mercury deposition in dated sediment cores reveal the impacts of the legacy and present-day human activities in Todosos Santos Bay, North-east Brazil. *Marine Pollution Bulletin*, 2019, 145, 396–406.
- [42] Ahmad, N.I., Mohd Fairulnizal Mohd, N., Wan Rozita Wan, M., Hamdan, J., Ismail, I., Wan Nurul Farah Wan, A., ..., Mohd Hairulhisam, H. Mercury levels of marine fish commonly consumed in Peni-

- nsular Malaysia. *Environmental Science and Pollution Research*, 2015, 22(5), 3672–3686.
- [43] Kehrig, H.A., Seixas, T.G., Baêta, A.P., Malm, O., Moreira, I. Inorganic and methylmercury: Do they transfer along a tropical coastal food web? *Marine Pollution Bulletin*, 2010, 60(12), 2350–2356.
- [44] Bjørklund, G., Aaseth, J., Ajsuvakova, O.P., Nikonorov, A.A., Skalny, A.V., Skalnaya, M.G., Tinkov, A.A. Molecular interaction between mercury and selenium in neurotoxicity. *Coordination Chemistry Reviews*, 2017, 332, 30–37.
- [45] Dyrssen, D., Wedborg, M. The sulphur-mercury (II) system in natural waters. *Water, Air, and Soil Pollution*, 1991, 56(1), 507–519.