

## Health Risk Assessment of Cadmium and Mercury via Seafood Consumption in Coastal Area of Nai Thung, Nakhon Si Thammarat Province, Thailand

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### Abstract

Metal contamination in seafood may cause adverse effects on human health. The objectives of this study were to determine the concentrations of cadmium and mercury in seafood from the coastal area of Nai Thung sub-district, Nakhon Si Thammarat Province, Thailand, and to evaluate the potential health risk of seafood consumption. A total of 46 samples (including 26 species of fish and shellfish) were caught in March and July 2018. Results indicated that the concentrations of metals in seafood samples were within the standards established for human consumption. However, cadmium concentrations in most shellfish samples were higher than the fish samples, and mercury in fish was higher than other marine organisms. The health risk assessment (HRA) of cadmium and mercury in seafood consumption was between 0.0004 - 0.07 and 0.001 - 0.035, respectively. The results showed that seafood from Nai Thung was unlikely to affect human health.

**Keywords:** Health risk assessment, Cadmium, Mercury, Seafood, Coastal, Nakhon Si Thammarat, Gulf of Thailand

### Introduction

Cadmium and mercury are in the top 10 primary public health concerns in the world [1]. They are commonly found in seafood, which is usually consumed by most people [2,3]. Therefore, the WHO/FAO (2011) recommends guidelines for provisional tolerable monthly intake (PTMI) of cadmium to be 25 µg/kg body weight and provisional tolerable weekly intake (PTWI) of mercury to be 4 µg/kg body weight. The contamination food standard of Thailand for mercury is 0.5 mg/kg [4]. Cadmium and mercury can be released into the environment by natural and human activities. The two primary sources of contamination are the production and utilization and the disposal of wastes containing heavy metals. They can be accumulated in aquatic organisms and transfer through the food chain, which leads to biomagnification [5,6]. The high contribution of cadmium and mercury to human exposures is shellfish and fish consumption [7-9]. The human kidney and bone are the main target organs for cadmium accumulation and effects [10]. The very high mercury concentrations cause damage to the nervous, digestive, and immune systems, lungs, and kidneys [11].

Thailand has 2,614 km of tropical coastline from the westerly Andaman Sea to the easterly Gulf of Thailand. The gulf is divided into two parts; The upper gulf is known for its intensive development and exploitation in major industry, tourism, and shipping, while the lower gulf is for oil and gas platforms, tourism, and fishery. Nevertheless, the coastal environment has been heavily exploited, causing adverse impacts as a result of the lack of comprehensive control focusing on environmental pressures, problems, and threats from the development [12]. Consequently, heavy metal contamination has been a major

pollution problem in the Gulf of Thailand [13] since the Gulf of Thailand is an important seafood source. Heavy metal contamination in marine organisms from oral exposures, including Cd, Cr, Cu, Hg, Pb, and Zn, has also been a major concern. Worakhunpiset reviewed heavy metals in marine organisms and sediment in the Gulf of Thailand and found that most of the articles pointed to the Upper Gulf of Thailand [14] as a place with heavy metal contamination. In the Lower Gulf of Thailand, heavy metal contamination in the environment was limited [13]. Some researchers reported concentrations of arsenic, cadmium, chromium, copper, lead, manganese, nickel, and zinc. It found that cadmium concentration in the Lower marine organism was higher than the other parts of the gulf, and arsenic and lead in sediment slightly higher than standard [15-19].

Seafood was used as a biological indicator to assess human health's adverse effects from consuming contaminated marine organisms, especially commercially food sources such as fish, crabs, and mollusk [8,9,20]. Since cadmium and mercury are toxic and biomagnification in aquatic organisms, the heavy metals in seafood were used to study HRA as an early warning for marine environmental assessments and seafood safety. In previous studies, a number of approaches have been proposed to assess the potential health risks of toxic metals exposure, and the target hazard quotient (the ratio between the dose of toxic metal exposure and the reference dose) has been generally applied in HRA in food [20-23]. Subsequently, the cadmium and mercury in the Lower Gulf have to be investigated to estimate the Gulf of Thailand's overall contamination situation and seafood concentrations from Nai Thung village, Nakhon Si Thammarat province, Thailand. This is also to evaluate the heavy metals for risk on seafood consumption.

## Materials and methods

### Study area

The study area is situated in the southern part of the Gulf of Thailand in South-East Asia. Nai Thung village is located Tha Sala district of Nakhon Si Thammarat province (**Figure 1**). Since 2012, Tha Sala Administration Organization is the 1<sup>st</sup> local authority in Thailand that enhances a local regulation on coastal conservation area including Nai Thung village. This village was chosen because of the strength of the local community organization called "Srabua-Naithung Fisher Folk Conservation Group". The organization is a great ally and local champion of neighboring villages to engage other key stakeholders to promote national and international programs on fishery conservation activities. The sampling sites were at 8°38'26.9"N 99°59'22.6"E, 8°38'08.6"N 99°59'14.1"E, and 8°38'17.2"N 99°59'23.7"E.



**Figure 1** Study area in the coastal fishing area of Tha Sala district, Nakhon Si Thammarat province.

### Sample collection

Various seafood, including 19 species of fish, one species of mollusk, and six species of crustacean, were collected in March and July 2018 at Nai Thung village. These were preferred as they are the most consumed by the local villagers and in Thailand. Moreover, they are present in the fishery throughout the year. Samples were collected in the accessible pools for the same species and were packaged in labeled plastic bags. They were cooled in the field by dry ice and transported to the laboratory at Walailak University. Subsequently, samples were measured for body length and then frozen at  $-20^{\circ}\text{C}$  until dissection. For small species, the entire edible parts of individuals were included when preparing composite samples, whereas, for large species, fillets of the edible parts of individuals were used. Samples were prepared for three replicates of each species for each sampling. Then, samples were weighed before and then freeze-dried for 24 hrs. to calculate the water content in tissues and the concentrations of metals in wet weight tissues.

### Heavy metal analysis

The dried samples of 0.25 g were weighed and digested with a mixture of highly purified concentrated 69 %  $\text{HNO}_3$ : hydrogen peroxide (5:2) at  $110^{\circ}\text{C}$  until the color of the solutions were pale yellow and clear. After the completion of the digestion and adequate cooling, solutions were filtered and made up to 50 mL with deionized water. The Cd concentrations were determined by an Inductively Coupled Plasma Optical Emission Spectrometer (Optima 8000; Perkin Elmer Instruments, USA). For Hg, the concentrations were determined by a Direct Mercury Analyzer (NIC MA-3000) using a Thermal Decomposition-Atomic Absorption Spectrophotometer. The limits of detection (LOD) were the concentrations which the instrument was able to detect and were calculated as 3 times of the standard deviation of blanks for Cd and Hg, which were 0.015 mg/kg and 0.5  $\mu\text{g/kg}$ , respectively. The limits of quantitation (LOQ) were the concentrations which the instrument was able to detect and quantify and were calculated as 10 times of the standard deviation of blanks. The LOQ for Cd and Hg the values were 0.05 mg/kg and 2.5  $\mu\text{g/kg}$ , respectively.

### Health risk assessment

Health risk assessments (HRA) of Cd and Hg were characterized using a hazard quotient, which is the ratio of the Chronic Daily Intake (CDI) divided by the oral Reference Dose of Cd and Hg. If the value is more than 1, the human health may be affected by metal accumulations in seafood [24].

The oral Reference Dose (RfD) assumes that thresholds exist for certain toxic effects. In general, the RfD is an estimate of a daily exposure to the human population that is unlikely to be affected during a human lifetime [25]. The RfD of Cd and Hg are 0.001 and 0.0001 mg/kg-day respectively [26,27]. The CDI was estimated by the following equation [24];

$$\text{CDI} = \frac{\text{Cs} \times \text{EF} \times \text{ED} \times \text{IR}}{\text{AT} \times \text{LT} \times \text{BW}} \quad (1)$$

Where

Cs = Concentration of seafood

EF = Exposure frequency (350 days/year)

ED = Exposure duration (years)

IR = Ingestion rate (seafood consumption rate; mg/day)

AT = Averaging time ( $365 \times \text{ED}$ ; days/years)

LT = Lifetime (equal to exposure duration; years)

BW = Body weight (kg)

The data of exposure duration, body weight, and ingestion rate of seafood were used from National Bureau Agricultural Commodity and Food Standards [28] as shown in **Table 1**.

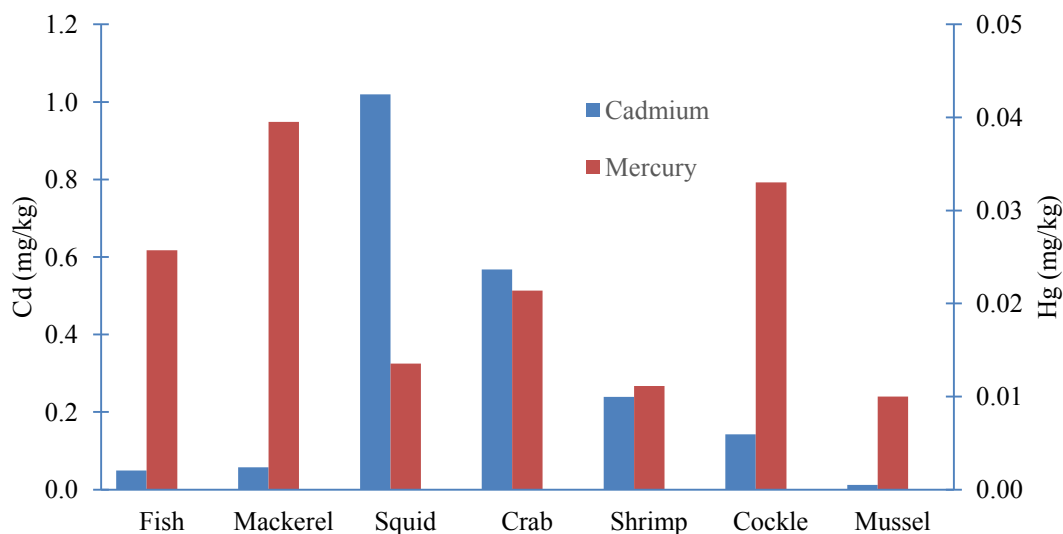
**Table 1** Consumption rate of seafood by the Thai population.

Range of ages (years)	Exposure duration (years)	Body weight (kg)			Ingestion rate (mg/kg-day)					
		Male	Female	Fish	Mackerel	Squid	Crab	Shrimp	Cockle	Mussel
3.0 - 5.9	6	17.5	17.0	0.69	6.39	2.80	0.70	3.95	0.97	0.59
6.0 - 12.9	12	32.6	34.3	0.79	7.34	4.61	0.82	5.20	1.71	0.92
13.0 - 17.9	18	56.3	50.8	0.71	7.97	5.31	0.81	5.14	1.82	0.83
18.0 - 34.9	35	67.1	59.6	1.70	9.21	5.42	1.19	4.55	2.06	1.46
35.0 - 64.9	65	65.9	61.8	1.45	8.96	2.36	0.86	2.65	1.02	0.79
> 65.0	74	56.9	53.1	0.88	6.16	0.89	0.27	1.50	0.32	0.27
Average	74	58.5	54.1	1.04	7.67	3.57	0.78	3.83	1.32	0.81

## Results and discussion

### Concentration of cadmium and mercury in seafood

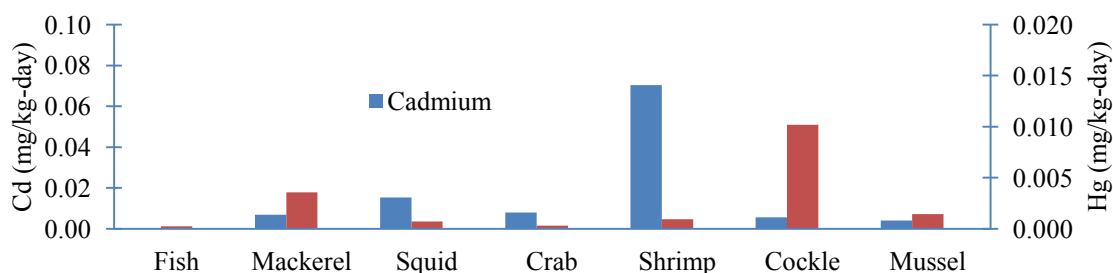
World Health Organization (WHO) and Food Agriculture Organization (FAO) established cadmium in food standards and divided seafood into crustaceans, mollusks, and other food. Cadmium concentrations in the food standard should be less than 0.5 mg/kg and 2.0 mg/kg for marine bivalve mollusks and Cephalopods. The concentrations of cadmium were highest in shrimp and lowest in fish from Nai Thung coastal areas. In **Figure 2**, cadmium concentrations in fish and crustaceans were in the range 0.012 - 1.060 mg/kg, which were lower than the food standards recommended by WHO/FAO (2011a and 2011c) [29,30]. Cadmium concentrations in seafood from Thailand accumulated in sediment more than in water [31]. Therefore, concentrations in benthic organisms such as crustaceans and mollusks were higher than in fish [15,32]. However, cadmium in food was not identified by the Ministry of Public Health in Thailand. For mercury, the concentrations were 0.011 - 0.04 mg/kg, which was lower than the food standard of Thailand and FAO/WHO (0.5 mg/kg) [4,30]. The highest concentration was found in fish, but the lowest concentration was found in squid. Because mercury in fish was transformed into methyl mercury in fish, it was biomagnification in the food chain. As fish are near the top layer of the marine food chain, it accumulates mercury more than other marine organisms [29,30]. In this study, shrimp and mussels were inappropriate to use as a bioindicator for mercury due to the least concentrations were found. Mercury concentrations in mussels were not detected in the Black Sea [33]. The concentrations of cadmium and mercury in seafood from the contaminated area in the Upper Gulf of Thailand were 0.0004 - 0.224 and 0.0005 - 5.23 mg/kg, respectively [23]. These results found that the heavy metals concentrations in the Lower Gulf of Thailand were lower than in the Upper Gulf of Thailand.



**Figure 2** Concentrations of cadmium and mercury in seafood from Nai Thung coastal area.

#### Chronic daily intake of cadmium and mercury in seafood

Chronic daily intake of seafood of cadmium and mercury were high in shrimp and cockle. Cadmium daily intake was less than 0.07 mg/kg-day, which was higher than the daily intake of mercury. Metals exposure to human from seafood consumption in Nai Tung village is lower than FAO/WHO standard and other countries because it is from a non-polluted area. Joint FAO/WHO Food Standard Programme CODEX Committee on Contaminants in Foods identified the PTMI for cadmium and mercury as 25 and 4  $\mu\text{g/kg}$  body weight, respectively [34]. It was applied to the CDI of seafood for cadmium and mercury as 0.171 and 0.075  $\mu\text{g/kg day}^{-1}$  respectively (**Table 2**). The highest dietary exposure of cadmium and mercury was found in Japan and China [29,35]. However, metal concentrations found in this study tend to be lower than in Asia, Europe, and continental America. Human exposures to metals were from varieties of food. This study showed that seafood consumption was not a major source for cadmium and mercury exposure. In Thailand, the major source of dietary Cd exposure are pig kidney and rice, which has a daily exposure of 0.03 and 0.06  $\mu\text{g/kg-day}$  [31,36].



**Figure 3** Chronic daily intake of cadmium and mercury from seafood consumption in the Nai Thung coastal area.

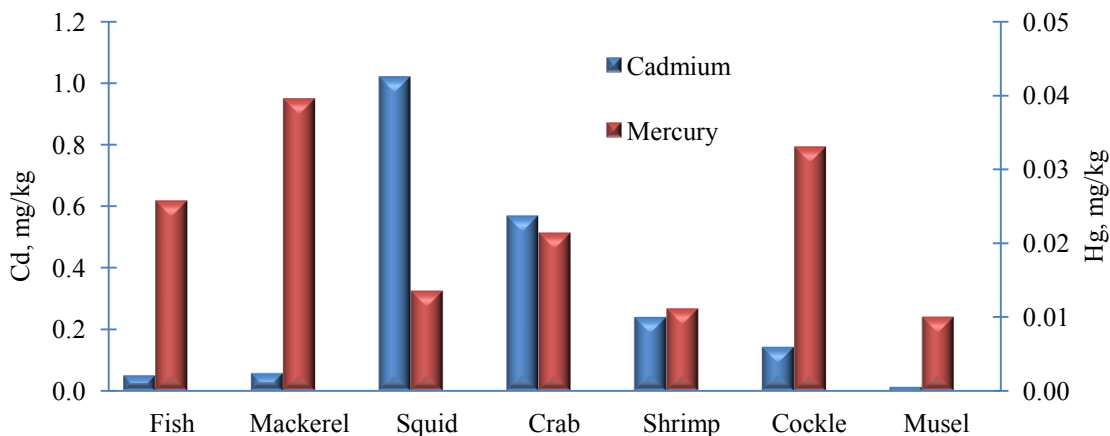
**Table 2** International estimates of daily exposure to cadmium and mercury for adults [30,34].

Country	Cadmium ( $\mu\text{g/kg-day}$ )	Mercury ( $\mu\text{g/kg-day}$ )
Australia	0.07	0.01 - 0.02
Chile	0.30	0.02
China	0.33	0.41 - 0.87
Europe	0.30	0.02 - 0.04
Japan	0.40	0.16
Korea	0.26	0.03
USA	0.15	0.01 - 0.02
Thailand (This study)	0.00 - 0.07	0.00 - 0.01
FAO/WHO Guideline	0.171	0.075

#### Health risk assessment of cadmium and mercury in seafood

The most important aspect to consider in seafood consumption is investigating the toxicity of metals to humans. Different approaches for the estimation of human health risks of metals in food have been proposed. The most widely applied comparison is the Provision tolerable weekly intake [23] or Provision tolerable monthly intake [31], representing the amount of substance that can be ingested over a lifetime without appreciative risks. Another way of determining risk is the HRA value proposed by the US.EPA. It is an integrated risk index that compares the ingested amount of a contaminant with a standard reference dose. A health risk assessment less than 1 indicates that the level of exposure is lower than the reference dose, which accepts that a daily exposure at this level is not likely to cause any adverse health effects during a lifetime in a human population [24].

Health risk assessments of cadmium and mercury in seafood from the Nai Thung coastal area showed variation for all species (**Figure 4**). Cadmium risk assessments for human health were in the range of 0.0004 - 0.070. The risk from cadmium was lower than mercury, excluding squid and crab (**Figure 5**). This was a result of the reference dose value of mercury being lower than cadmium [26,27]. The highest risk of cadmium was found in shrimp because the concentrations of cadmium in tissues were high (**Figure 2**). It is apparent from these results that the health risk of cadmium is lower than in other countries in the world except Turkey (**Table 3**). For mercury, the HRA was 0.001 - 0.036. The highest risk for mercury was found in mackerel due to seafood's highest consumption rate for the Thai population (**Table 1**). The health risk value of mercury in the study area was lower than in other countries and a polluted area in the Gulf of Thailand (**Table 3**). Although the present calculations were less than 1, the chemical forms of mercury in seafood should be investigated because the toxicity of Hg in organic forms, such as methyl mercury, are the most toxic ones [37]. Moreover, it found that the health risk from metal concentrations in this study area was lower than in polluted areas in the Gulf of Thailand. Thongra-ar *et al.* [23] showed that the risk of cadmium and mercury in seafood was in the range 0.0 - 3.94 and 0.01 - 1.06, respectively. The highest risks of cadmium and mercury were found in scallops and sand whiting.



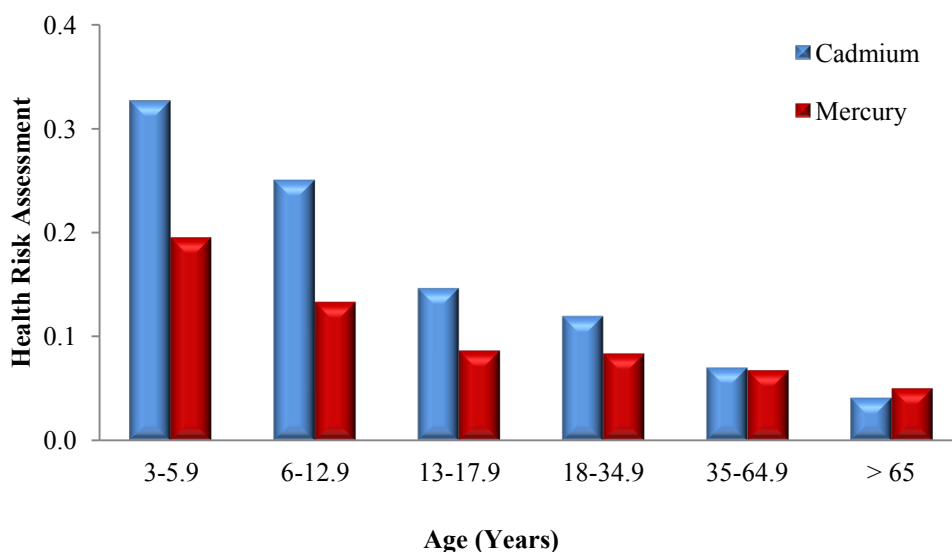
**Figure 4** Human health risk assessment of cadmium and mercury for seafood consumption in Nai Thung coastal area.

**Table 3** Human health risk assessment of cadmium and mercury in the world.

Areas	Cadmium	Mercury	References
Turkey	0.0 - 0.01	0.13 - 3.19	Mol <i>et al.</i> , 2018 [38].
Caribbean	N.A.	$0.54 \pm 0.23$	Fuentes-Gandara <i>et al.</i> , 2018 [22].
Peru	0.0 - 1.25	N.A.	Loaiza <i>et al.</i> , 2018 [39].
Bangladesh	0.0 - 2.40	0.0 - 0.07	Baki <i>et al.</i> , 2018 [40].
Italy	0.0 - 0.35	0.0 - 0.15	Barone <i>et al.</i> , 2015 [37].
Upper Gulf of Thailand	0.0 - 3.94	0.01 - 1.06	Thongra-ar <i>et al.</i> , 2014 [23].
	0.002 - 0.013	0.001 - 0.002	Sudsandee <i>et al.</i> , 2017 [41].
Lower Gulf of Thailand	0.0004 - 0.07	0.001 - 0.036	This study

Note: N.A. means not available.

Moreover, the results showed that human health risk of cadmium and mercury in children were higher than adult (**Figure 4**), it was because the body weight and the seafood consumptions of children was lower than adult. Gender also influenced the value of HRA, as female was higher than male. It is apparent from these results that the metals exposure health risk is associated with the body weight of the population. This is a similar scenario to that in China where children were more likely to be affected by heavy metal pollution than adults [42].



**Figure 5** Health risk assessment of cadmium and mercury for Thai individuals in various age groups.

## Conclusions

In the present study, seafood samples including fish, mackerel, squids, shrimp, and bivalves were collected and analyzed for cadmium and mercury concentration. The risk assessment of cadmium and mercury for seafood consumption was less than 1. Consequently, cadmium and dietary mercury exposure for seafood consumption do not seem to pose any hazards to human health. Moreover, the heavy metal concentrations did not exceed the Ministry of Public Health's permission limit of Thailand and WHO/FAO guidelines. Analytical data showed significant differences in metal concentrations among the different species. Cadmium concentrations were high in shrimp and mussel, and mercury concentrations were high in mackerel. The results suggest caution in seafood consumption, especially in shellfish and fish and particularly population segments such as young children. It may be suggested that continuous biomonitoring must be taken to heavy metal concentrations in seafood.

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