

## Estimation of Target Hazard Quotients and Potential Health Risks for Metals by Consumption of Shrimp (*Litopenaeus vannamei*) in Selangor, Malaysia

(Penganggaran Darjah Bahaya Sasaran dan Potensi Risiko Kesehatan untuk Logam melalui Pengambilan Udang (*Litopenaeus vannamei*) di Selangor, Malaysia)

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### ABSTRACT

*The concentrations of the heavy metal in Pacific white shrimp (Litopenaeus vannamei) purchased from the local wet markets throughout the state of Selangor were investigated using flame atomic absorption spectrometry. The order of the heavy metal concentrations were Zn>Cu>Pb>Cd, whereby the metal concentrations in most samples exceeded the limits of the Malaysian Food Regulation, 1985 and the guidelines set by the Food and Agriculture Organisation (1983). A health risk analysis based on the mean target hazard quotient (THQ) yielded values <1 for all metals for average and maximal consumers. The total THQ (tTHQ) which measures the aggregated risk due to heavy metal uptake via the ingestion of L. vannamei was 0.124 and 0.372 for average and maximal consumers, respectively. This suggests that although the metal concentrations are exceeding the limit, there is no calculated significant risk from metal toxicity by the consumption of shrimp.*

*Keywords: Hazard quotient; health risk assessment; heavy metals; shrimp*

### ABSTRAK

*Kepekatan logam berat dalam udang putih (Litopenaeus vannamei) yang dibeli dari dua puluh pasar basah di negeri Selangor telah diukur dengan menggunakan spektrometri penyerapan api atom. Purata tahap kepekatan logam berat adalah Zn>Cu>Pb>Cd, dengan kebanyakan sampel melebihi had Peraturan Makanan Malaysia, 1985 dan garis panduan yang ditetapkan oleh FAO (1983). Analisis risiko kesihatan berdasarkan darjah bahaya sasaran (THQ) menghasilkan nilai <1 untuk semua logam untuk pengguna. Jumlah THQ (tTHQ) yang mengukur risiko agregat disebabkan oleh pengambilan logam berat melalui pemakanan L. vannamei masing-masing adalah 0.124 dan 0.372, untuk pengguna yang makan udang secara sederhana dan maksimal. Ini mencadangkan walaupun kepekatan logam berat melebihi had yang ditetapkan, tiada risiko ketara daripada ketoksikan logam melalui pengambilan udang.*

*Kata kunci: Darjah bahaya; logam berat; penilaian risiko kesihatan; udang*

### INTRODUCTION

The Pacific white shrimp (*Litopenaeus vannamei*), locally known as “udang putih” lives in tropical marine areas where water temperature exceeds 20°C throughout the year. The global production of Pacific white shrimp was 3,220,037 tons in 2012 due to rapid spread of the species to Asia; additionally, Malaysia is one of the main producers of this species (FAO 2015). Shrimp is a common food consumed by humans and supplies a good source of high quality protein, vitamins and dietary minerals (Baboli et al. 2013). However, food of marine origin, albeit excellent sources of essential micronutrients may contain zinc, iron, copper, mercury, manganese, cadmium, lead and arsenic absorbed from the environment (Belitz et al. 2001; Fernandes et al. 2008).

In marine water, high concentrations of heavy metals can enter to the environment coastline (Mitra et al. 2010) usually sourced from dissolved and solid waste from industrial and agricultural activities transported

into aquatic habitats (Enuneku et al. 2014). There is significant industrial pollution in the Straits of Malacca (Chua et al. 2000), whereby many studies have found high concentrations of metals in these waters (Abdullah et al. 1997; Chua et al. 1997; Ismail 1993; Sivalingam & Bhaskaran 1980; Yap et al. 2004).

Biomagnification of heavy metals from the environment into edible shrimp is a cause for concern as humans are at the highest level in the trophic level of the food chain; therefore, it often becomes mandatory to check chemical contaminants in foods from the aquatic environment to understand their hazard levels. The majority of people obtain their shrimp from fish markets and supermarkets (Burger et al. 2005), making it important to know the level of contaminants. Therefore, this study aimed to assess the level of risk posed to the population by copper, cadmium, lead and zinc in shrimp purchased from local wet markets in Selangor.

## METHODS

### FIELD SAMPLING

The sampling period spanned from March-May 2015. Specimens of *L. vannamei* were purchased randomly twice from 20 local markets in Selangor. The samples were put in pre-cleaned polythene bags and preserved in ice chest at -4°C and transferred immediately to the laboratory. The samples are stored at -18°C before analysis and the steps were repeated for other sampling points.

### SAMPLE PREPARATION AND ANALYSIS

Samples were initially dried in the oven at 80°C for 24 h prior to acid digestion (Hashmi et al. 2002). Two grams of dried sample were weighed with a digital weighing scale and digested in 10 mL concentrated HNO<sub>3</sub> at 60°C for 30 min. The samples were cooled to room temperature and 2 mL of hydrogen peroxide was added. The solution was again heated in a digester until brown fumes were expelled, tissues dissolved completely and a colourless solution was obtained. The contents were allowed to cool and thereafter the digested sample was made up to 100 mL with distilled water. AA Analyst 400 Perkin Elmer Atomic Absorption Spectrometer (AAS) was used for the analysis of heavy metals in shrimp.

### CHRONIC DAILY INTAKE (CDI) OF METALS

The chronic daily intake (CDI) of elements is dependent on both the concentrations in shrimp and the amount of shrimp consumed (USEPA 2000). The average shrimp consumption rate is 3 g/day (MOH 2006). However, accounting for maximally exposed individuals consuming a weekly shrimp meal serving, the maximum consumption rate is 9 g/day.

$$CDI = \frac{IR \times C}{BW} \quad (1)$$

where IR is the food ingestion rate (g/day); C is the heavy metal concentration in shrimp (µg/g); and BW is the average adult body weight (60 kg).

### RISK ASSESSMENT

The estimation of non-carcinogenic health hazards from the consumption of shrimp was determined by the equation provided in the United States EPA Region III Risk based concentration table (USEPA 1989).

The target health quotient (THQ) is:

$$THQ = \frac{EFr \times ED \times IR \times C \times 10^{-3}}{RfD \times BW \times AT} \quad (2)$$

where EFr is exposure frequency (365 days/year); ED is the exposure duration 70 years (average lifetime); IR is the food ingestion rate (g/day); C is the heavy metal concentration

in shrimp (µg/g); RfD is the oral reference dose (mg/kg/day); BW is the average adult body weight (60 kg); and AT is the averaging exposure time for non-carcinogens (365 days/year × number of exposure years). The applied RfD for copper, cadmium, lead and zinc was 0.04, 0.001, 0.004, 0.3 mg/kg/d, respectively (USEPA 2007). Exposure to multiple pollutants may cause interactive or additive effect; hence, the total THQ (tTHQ) was calculated as the sum of each individual metal THQ for each sampling location. A tTHQ <1 means that the exposed population is assumed to be safe; however, when the tTHQ is between 1 and 5 there is a potential risk related to the studied metal in the exposed population (USEPA 2010).

### STATISTICAL ANALYSIS

Interstation comparisons were carried out to test for significant differences in the concentration of the heavy metals in the shrimp samples using parametric analysis of variance (ANOVA).

## DATA ANALYSIS AND RESULTS

### METAL CONCENTRATION IN SHRIMP

The concentration of cadmium in shrimp ranged from 0.22-3.08 mg/kg whereas lead concentration ranged between 2.25-4.87 mg/kg. Copper and zinc concentrations were much higher than the other metal concentrations and ranged between 12.80-48.50 and 39.96-68.38 mg/kg, respectively. Forty percent of the samples were above the permissible level for cadmium whereas for lead all samples were in violation of the Malaysian standards. The Malaysian Food Act 1983 (Act 281), Food Regulation 1985 sets the limit for cadmium and lead at a maximum of 1 mg/kg. The Malaysian legislation does not provide for the concentration of copper and zinc in shrimp, hence the suggested limits of 30 mg/kg by the Food and Agricultural Organization (1983) is used. All samples were above the limit for zinc concentration while 40% of the samples had a copper concentration of above 30 mg/kg. The metals characterized in the shrimp samples are summarized in Table 1. All metal concentration showed significant difference ( $p > 0.05$ ) among the twenty designated sampling locations.

### CHRONIC DAILY INTAKE (CDI)

The chronic daily intake (CDI) of heavy metals was evaluated according to the concentration of each metal in shrimp and the respective consumption rate. The CDI of the studied metals from consumption of shrimp are shown in Table 2. In shrimp samples, mean values of CDI showed the descending order of zinc > copper > lead > cadmium. The CDI of the studied metals through consumption of shrimp were lower than the tolerable intake suggested by the Joint FAO/WHO Expert Committee on Food Additives (JEFCA 2009) (Table 2).

TABLE 1. Variation in metal concentration (mg/kg) in shrimp

Location	Metal concentrations (mg/kg); Mean (SD)			
	Copper	Cadmium	Lead	Zinc
W1	17.35 (0.68)	3.08 (0.21)	2.31 (0.02)	44.50 (4.31)
W2	18.34 (0.01)	0.97 (0.11)	2.25 (0.09)	49.33 (1.10)
W3	19.59 (1.01)	0.79 (0.01)	2.27 (0.15)	39.96 (1.05)
W4	33.12 (1.28)	1.00 (0.04)	2.10 (0.04)	49.10 (5.97)
W5	19.38 (0.81)	0.98 (0.06)	2.40 (0.38)	55.41 (3.97)
W6	18.22 (0.04)	0.99 (0.07)	2.91 (0.05)	51.52 (1.96)
W7	20.89 (0.90)	1.01 (0.04)	2.81 (0.18)	50.75 (5.92)
W8	17.63 (0.32)	1.15 (0.02)	4.87 (0.33)	64.39 (5.21)
W9	12.80 (0.59)	1.76 (0.06)	3.03 (0.08)	68.04 (10.63)
W10	26.09 (0.99)	1.17 (0.05)	2.83 (0.01)	60.35 (11.55)
W11	48.50 (3.38)	0.81 (0.04)	2.90 (0.06)	61.31 (9.00)
W12	45.06 (1.49)	1.60 (0.10)	2.69 (0.14)	76.99 (3.40)
W13	20.13 (0.50)	0.24 (0.01)	2.61 (0.01)	71.95 (1.20)
W14	27.75 (1.07)	0.48 (0.02)	2.45 (0.05)	67.85 (7.16)
W15	34.83 (0.66)	0.22(0.02)	2.36 (0.05)	70.54 (5.67)
W16	23.74 (0.00)	0.37 (0.17)	2.43 (0.09)	66.50 (4.25)
W17	16.17 (0.63)	0.48 (0.02)	2.64 (0.03)	75.44 (3.74)
W18	25.45 (2.16)	0.74 (0.07)	2.46 (0.24)	68.38 (12.60)
W19	20.80 (0.34)	1.32 (0.12)	2.71 (0.04)	70.87 (1.72)
W20	36.18 (2.48)	0.43 (0.02)	2.47 (0.01)	74.60 (1.00)

Values in shaded cells are in violation of the standards

TABLE 2. Chronic daily intakes (CDI) (ug/kg BW/day) of metals by consuming shrimp

Location	average exposed consumer				maximally exposed consumer			
	Copper	Cadmium	Lead	Zinc	Copper	Cadmium	Lead	Zinc
W1	0.87	0.15	0.12	2.22	2.60	0.46	0.35	6.67
W2	0.92	0.05	0.11	2.47	2.75	0.15	0.34	7.40
W3	0.98	0.04	0.11	2.00	2.94	0.12	0.34	5.99
W4	1.66	0.05	0.10	2.46	4.97	0.15	0.31	7.37
W5	0.97	0.05	0.12	2.77	2.91	0.15	0.36	8.31
W6	0.91	0.05	0.15	2.58	2.73	0.15	0.44	7.73
W7	1.04	0.05	0.14	2.54	3.13	0.15	0.42	7.61
W8	0.88	0.06	0.24	3.22	2.64	0.17	0.73	9.66
W9	0.64	0.09	0.15	3.40	1.92	0.26	0.45	10.21
W10	1.30	0.06	0.14	3.02	3.91	0.18	0.42	9.05
W11	2.43	0.04	0.14	3.07	7.28	0.12	0.43	9.20
W12	2.25	0.08	0.13	3.85	6.76	0.24	0.40	11.55
W13	1.01	0.01	0.13	3.60	3.02	0.04	0.39	10.79
W14	1.39	0.02	0.12	3.39	4.16	0.07	0.37	10.18
W15	1.74	0.01	0.12	3.53	5.22	0.03	0.35	10.58
W16	1.19	0.02	0.12	3.33	3.56	0.06	0.36	9.98
W17	0.81	0.02	0.13	3.77	2.43	0.07	0.40	11.32
W18	1.27	0.04	0.12	3.42	3.82	0.11	0.37	10.26
W19	1.04	0.07	0.14	3.54	3.12	0.20	0.41	10.63
W20	1.81	0.02	0.12	3.73	5.43	0.07	0.37	11.19
MEAN	1.25	0.05	0.13	3.09	3.76	0.15	0.40	9.28
TOLERABLE INTAKE (ug/kg BW/day)					500	0.83	3.57	300-1000

## ESTIMATION OF POTENTIAL HEALTH RISK

Target hazard quotients (THQs) of the four studied metals from consuming shrimp are depicted in Figure 1. The THQ values for all metals in all locations were lower than 1, indicating no calculated health hazard to humans. The total THQ (tTHQ) values for average and maximally exposed consumers from all the studied metals were lower than 1 for all locations (Figure 2).

## DISCUSSION

## CONCENTRATIONS OF METALS

The various analysed heavy metals were found to have bioaccumulated in the whole tissue of *L. vannamei* sampled in the order of magnitude zinc>copper>lead>cadmium. Cadmium was the least accumulated heavy metal in shrimp sampled from all locations. Metal concentrations were relatively high with 66.2% of the samples exceeding the permissible limits. The highest readings were obtained from Location W11, Location W1, Location W8 and Location W12 for concentrations of copper, cadmium, lead and zinc, respectively. Aquatic organisms are commonly known for their ability to accumulate chemicals and considered to be a bioindicator for the marine environment (Yunus et al. 2012). The differences in the metal concentrations are attributable to the

prevailing anthropogenic activities going on at the aquatic ecosystems where the shrimp are sourced from. However, the exact source of contamination cannot be determined as the shrimp found in the wet markets are from different local sources. The shrimps are mostly bought wholesale either from local or interstate shrimp farms and sold to the consumers. In addition, some of the shrimp sellers also market wild shrimp caught from the Straits of Malacca. Fertilizers to encourage plankton growth (Hashim & Kathamuthu 2005) and biocides inserted into shrimp ponds before harvesting (Boyd 2002) may contribute to the presence of metals in the water. Additionally, estuaries and coastal areas where shrimps are farmed receive high anthropogenic inputs from both non-point and point sources (Mokhtar et al. 2009). Studies show that the high level of heavy metals in tissues of marine organisms correlate with the heavy metal concentrations in water and sediments (Vicente-Martorell et al. 2009).

## DIETARY EXPOSURE AND POTENTIAL HEALTH RISKS

The dietary exposure approach is a reliable tool for investigating a population's diet in terms of intake levels of nutrients, bioactive compounds and contaminants, providing important information about the potential nutritional efficiencies or exposure to food contaminants (WHO 1985). The CDI for all the metals analysed from all

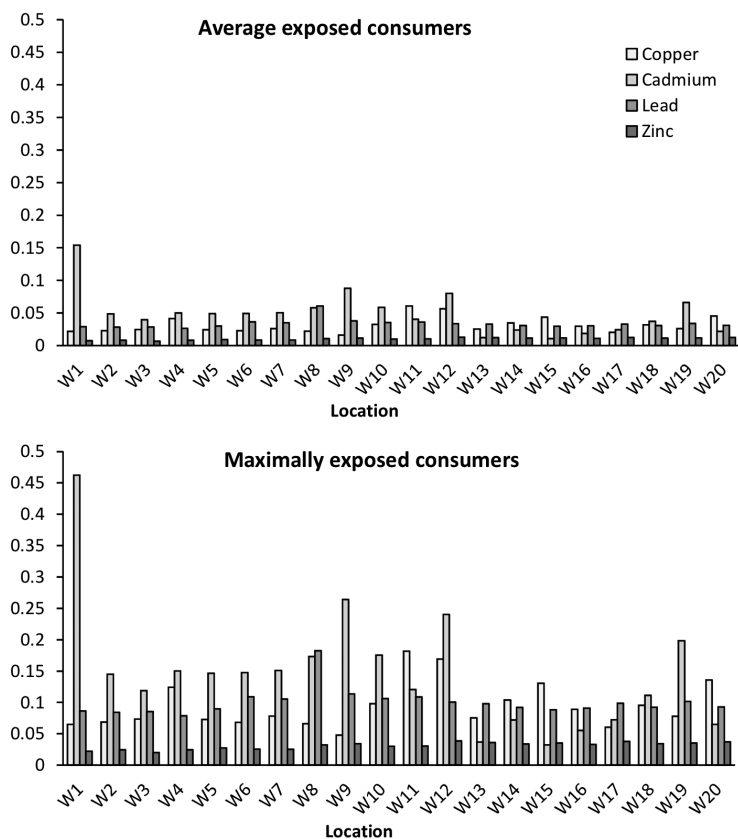


FIGURE 1. Target hazard quotient (THQ) by consuming shrimp of average and maximally exposed consumers

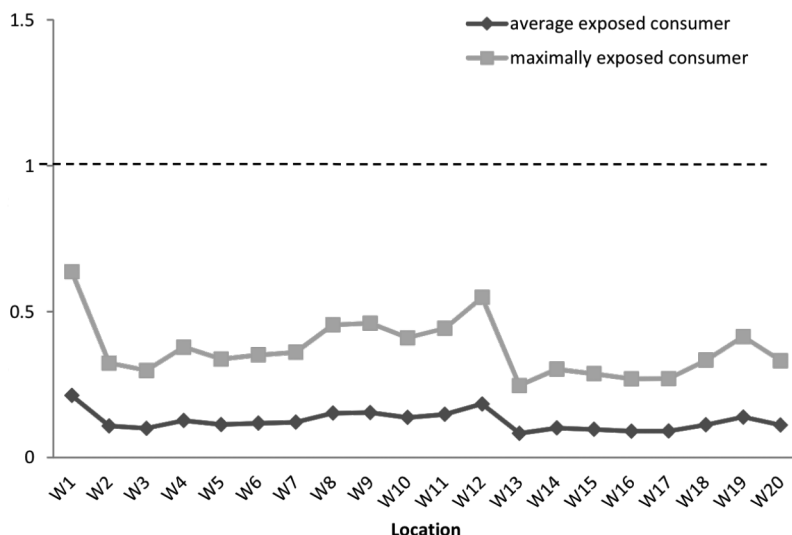


FIGURE 2. Total target hazard quotient (tTHQ) by consuming shrimp of average and maximally exposed consumers. The population will experience health risk if the tTHQ is equal or greater than 1

locations was well within the Tolerable Intake suggested by the Joint FAO/WHO Expert Committee on Food Additives (JEFCA 2009) and is not a health risk. Despite zinc having the highest concentration among the metals and all the lead and zinc concentration were in violations of the standards, the THQ value was below 1.00 for both zinc and lead. This is indicative that the presence of both these metals in shrimp does not pose a health risk to the consumers. Additionally, the THQ for copper and cadmium individually from all locations were also below 1.00 for both the average consumer (one meal per three weeks) and the maximally exposed consumer (one meal per week). However, potential health risks from exposure to shrimps are of some concern as different metals could have similar damage on some health endpoints. Therefore, the total THQ (tTHQ) is reliably helpful to assess and compare their combined risks and have been widely employed in recent literature (Song et al. 2009; Wang et al. 2005). The tTHQ ranged between 0.08 and 0.21 for the average exposed consumer and 0.24 and 0.63 for the maximally exposed consumer. THQ for individual metals were in general in the order zinc>copper>lead>cadmium, respectively, contributing 8.4, 25, 38.8 and 27.8% of the total THQ in the shrimp. This demonstrated relative cause for concern for lead and cadmium, as regular and daily consumption of shrimp may cause deleterious effects during a lifetime. Moreover, the exposure to lead and cadmium from other sources such as fish and organ meats is not calculated. The contribution to metal exposure depends largely on the consumption habits of the population and the divergent occurrence data found in food matrices. In addition, environmental pollution, especially in heavily industrialized areas, may contaminate aquaculture farms situated within the vicinity of related factories, where trace amounts of metals may be released from industrial processes and bioaccumulated in the body tissues.

## CONCLUSION

The main reasons the present study was developed was to provide baseline data on accumulation of toxic and essential metals in shrimp species from local wet markets in Selangor and to assess the potential health risks associated with consumption of shrimp. The investigated shrimp varied widely in their metal concentrations, and the estimated amounts of copper, cadmium, lead and zinc from shrimp were higher than the respective allowable concentration. The daily intake of metal and THQ suggest that the consumption of shrimp from locations in Selangor is free from risks in these areas. The complex total THQ parameters used in health risk assessment of heavy metals provides a better image than using only a simple parameter. However, individual and total target hazard quotient (tTHQ) would not pose any potential risk. It is thus suggested that constant monitoring of heavy metals is needed in all food commodities aiming to evaluate if any potential health risks from dietary metal exposure do exist. Nevertheless, it should be noted that limitation of the study was the relatively small sample size; hence these findings cannot be generalized for the nation. Additionally, the source of the shrimp is unknown and it could have been imported from neighbouring countries.

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