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Hair Mercury Levels in Relation to Marine Fish Consumption among Adults in Malaysia

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Abstract

Consumption of carnivorous fish is one of the major sources of human exposure to methyl mercury (MeHg). This study presents the data on fish consumption habits and hair mercury levels taken from 201 adults aged between 17- 72 years from four districts in the Peninsular Malaysia. The means for total mercury (THg) ranged from 0.93 ppm-1.69 ppm. The means for females and males were 1.21 ppm (SD=0.76) and 1.48 ppm (SD=0.89) respectively. The THg level for 59.30% of the study population (26.60% female participants) exceeded the USEPA recommendation of 1 ppm. The average fish consumption was 180.19±11.34g/ day/person with higher fish consumption in both rural coastal areas compared with the urban communities (*p= 0.0001*). Age was positively correlated with THg (r_s=0.4588; *p= 0.0001*) followed by the amount of fish eaten (r_s=0.4199; p=0.0001), use of whitening cream (r_s=0.2410; *p= 0.006*), BMI (r_s=0.2034; *p= 0.0041*), location of study (r_s=0.1818; *p= 0.01*), and gender (0.1637; *p= 0.0241*). However, we found negative correlation between the numbers of filling with THg (r_s=-0.2485; *p= 0.004*).

Keywords: fish consumption; THg; adult; hair mercury; Malaysia.

1. Introduction

Consumption of fish has been shown to have benefits as well as risks. Low levels of fish intake appeared to reduce chronic heart diseases and mortality. On the other hand, eating contaminated fish may pose the danger of heavy metal poisoning such as MeHg. In March 2004, the U.S. Food and Drug (FDA) and U.S. Environmental Protection Agency (USEPA) issued a joint advisory recommendation that pregnant women, young children and nursing mothers modify their fish consumption (Bouzan *et al*., 2005; Cohen *et al*., 2005).

Despite the Minamata tragedy that occurred in 1953 (Harada *et al*., 1999), some parts of the world are still facing mercury poisoning. Fifty-seven percent of the Amerindians in the French Guyana had mercury levels exceeding 10 ppm, associated with eating carnivorous fish with mercury levels up to 1.62 ppm (Frery *et al*., 2001). Extensive studies by Santos *et al*. (2002) showed that high fish consumption (especially carnivorous fish) was linked to elevated hair mercury level among Amazon natives. For the Faroe Islanders, eating pilot whale meat and blubber contributed to mercury exposure; where the meat contained 2-3 ppm mercury (Weihe and Gradjean, 2006).

What about Malaysia? Are we at risk of mercury exposure through fish consumption? Fish is an important food item which contributes to about two-thirds of all meat consumed in the country. Our annual fish consumption is as high as 57.7 kg/year (Laurenti, 2002). Over 90% of the national fish catch is from the sea (Mohd Arshad and Mohd Noh, 1994). Agusa *et al*. (2005) reported that some specimens of marine fish caught in the Straits of Malacca had mercury levels higher than the guideline value imposed by the USEPA. According to the Malaysian Environmental Quality Report (Department of Environment Malaysia, 2005), areas located in the northern part of the country have more than 10% observations of mercury levels exceeding the Malaysian Interim Marine Water Quality Standard of 0.001 mg/L (Table 1).

Currently, there are limited studies on THg in hair carried out in Malaysia. Table 2 summarizes the hair mercury levels for residents living in selected areas. In most study areas, the reported mercury levels exceeded the USEPA reference dose of 0.1 μg Hg/g hair which corresponds to the hair mercury of 1 ppm (Yasutake *et al*., 2003; Johnson *et al*., 2004).

Human hair has been considered as a valid bio-indicator to estimate human exposure to mercury (Airey

Table 1. Percentage of observations of mercury exceeding the Malaysian Marine Water Interim Standards for 7 selected location in Malaysia (1999-2004)

Location	% Observation for Parameter Hg Exceeding Interim Standards					
	1999	2000	2001	2002	2003	2004
Perlis	-	50	-	50	25	23
Langkawi	-	33	-	26	12	29
Kedah	-	58	-	50	0	27
Penang	-	34	-	20	13	10
Kelantan	-	0	-	0	0	0
Terengganu	-	2	-	0	18	1

1983; Matsubara and Michida 1985; Canuel *et al.*, 2006). In environmental exposure assessments, hair samples were used as biomarkers in assessing recent exposure to MeHg that occurred over the past several months or a year (Pinheiro *et al.*, 2005). Blood and urine on the other hand, measures very recent exposure (such as few days prior). The concentration of MeHg is approximately 250 times higher in hair than in blood. On average, the hair grows 1 cm per month (USEPA 1997); therefore, analysis of different sections of hair provide can information on temporal MeHg exposure.

This paper describes the pattern of fish consumption in relation to hair mercury levels among adults residing in the northern and north east states of the Peninsular Malaysia.

2. Materials and Methods

Between March to May 2006, 201 adults were recruited for hair mercury assessment. This study was carried out in collaboration with the National Institute for Minamata Disease (NIMD), Minamata, Japan, Kota Bharu Health Department, Bachok Health Department, Kedah Department of Environment, Kelantan Department of Environment and Kampung Balai Health Clinic. The proposal for this study had been reviewed and approved by the Ethics Committee, Universiti Putra Malaysia.

2.1. Study location

Two states, Kedah and Kelantan, were chosen for this study. Kedah is located northwest and Kelantan northeast of Peninsular Malaysia. In Kedah, two areas were chosen, Yan and Alor Setar. Yan, a fishing village, is located in the area is reported to have Hg levels exceeding the Malaysia interim marine water quality standard for mercury. Alor Setar, an urban area, is located 37 kilometers away from Yan. For Kelantan, Bachok, a fishing village, and Kota Bharu, an urban area 12 kilometers away from Kota Bharu, were chosen for this study.

2.2. Exposure assessment survey

With the help of a nurse and health inspectors, we sampled hair and distributed mercury exposure questionnaires to the participants. Each participant was interviewed using the questionnaire as a guide. The questionnaire contains information such as age, gender, race, weight, height, fish consumption (the quantity and frequencies of consumption), the number of amalgam dental fillings (if they had any), current or past occupations (such as gold mining, hospitals or laboratories, chemical or pesticide industries), the use of skin lightening-cream, traditional medicine, and, history of hair waiving.

The participants were also asked about their fish eating habits. Questions on fish consumption include: "How often do you eat fish?" The response was recorded as (1) *everyday* (_____ meals per day); (2) sometimes (a meal per _____ days); (3) rarely (a meal per _____ weeks or _____ months); and ( ) never (less than a meal per year). They were also asked to list the names of fish that they often consume.

Hairs were cut from at least three different areas of the scalp, about 3 cm (or less) from the hair root. The minimum amount required is twenty strands of hair, each 10 cm in length. Hair samples were put in a paper envelope, sealed and labeled for each participant. All samples were sent to the NIMD, Minamata, Japan. The THg levels were determined by oxygen combustion-gold amalgamation method using an atomic absorption detector MD-1. The limit of detection was 0.05 ng mercury.

2.3. Statistical Analysis

All statistical analyses were conducted with SAS, version 8 for Windows (SAS Inc., North Carolina).

Table 2. THg contents in scalp hair based on previous studies carried out in Malaysia

State / Race*	N	Range (or Highest level) (ppm)	Mean (ppm)	Median (ppm)	Reference
1) Penang	80	12.8 (<18 yr) 14.8 (19-40 yr) 16.1 (>41 yr)	7.36 7.90 9.19		Sivalingam and Sani (1980)
2) Selangor	35		4.3 (rural) 9.0 (urban)		Sarmani *et al*. (1984)
3) Kuala Lumpur	25	2.06 - 16.40	6.62 ± 3.73	7.03	
Sepang	20	1.21 - 7.97	4.34 ± 2.39	4.24	Sarmani (1987)
Alor Setar	40	0.20 - 1.52	0.82 ± 0.31	0.89	
Overall	85	0.20 - 16.40	3.36 ± 3.60 (AM) 2.97 ± 6.57 (GM)	1.24	
4) Penang	106	0.45 - 16.87	3.61 (AM) 3.49 (GM)	2.96	
Trengganu	33	6.79 - 18.31	12.08 (AM) 11.69 (GM)	12.05	Sarmani *et al*. (1994)
Selangor	45	0.66 - 6.90	3.01 (AM) 2.95 (GM)		
5) Malay		0.83 - 15.30	3.07	2.75	Sarmani *et al*.
Indian		0.59 - 18.73	4.43	3.81	(2002), Sarmani
Chinese Overall		0.91 - 15.69	5.12	4.50	and Alakili (2004a)
Kuala Lumpur	400	0.59 - 18.73	4.01	3.38	
6) Malaysians	400	0.59 - 18.73		3.38	Sarmani and
Libyan	50	0.00 - 3.60		0.81	Alakili (2004b)
Jordan	22	0.32 - 4.00		0.69	
7) Johor	46	0.60 - 19.76	9.94		
Urban		0.60 - 19.76	9.84		
Rural		3.80 - 17.40	10.31		
Terengganu	132	0.10 - 19.90	10.85		
Urban		0.98 - 19.90	9.82		
Rural		0.10 - 19.75	12.47		
Kedah	153	0.05 - 21.00	13.69		Hajeb *et al*. (2008)
Urban		0.05 - 20.50	11.41		
Rural		3.36 - 21.00	15.99		
Selangor	50	0.02 - 19.74	6.78		
Urban		0.02 - 17.29	5.34		
Rural		0.38 - 19.74	8.22		

Spearman's Correlation test is used to test for association between variables. The difference between groups is tested using the Wilcoxon rank-sum and Kruskal-Wallis tests.

3. Results

3.1. Demographic characteristics

Two hundred and one people participated in this study, consisting of 50.75% (102/201) female and 49.25 % (99/201) male. The participants' ages ranged from 18 to 72 years old (mean=40.6). The mean ages for participants in Yan (Kedah) and Alor Setar (Kedah) are 47.3 and 35.1 years old respectively, while for Bachok (Kelantan) and Kota Bharu (Kelantan) the ages are 43.7 and 36.3 years old respectively. Majority of the study population (64.7%) had annual household income below RM10,000. All participants from Yan and Bachok were fishermen whereas those from Alor Setar and Kota Baharu were mostly office workers.

3.2. Mercury level in hair

To monitor THg exposure in individuals, hair samples were taken. Descriptive statistics for THg according to gender and location is shown in Table 3. Our data showed that females from Yan exhibited the lowest THg (mean 0.98 ppm, SD=0.33) while the highest was for males from Yan (1.92 ppm, SD=1.06). The males had higher THg level in comparison to females. The mean for males 1.48 ppm (SD=0.89) and females was 1.21 ppm (SD=0.76). The mean for all participants was 1.34 ppm (SD=0.840 ppm). Two individuals, both female, who recorded high THg (223.58 and 803.16 ppm) were excluded from this analysis. Although their THg was high, the percentages of MeHg compared to THg were only 0.61% and 0.24% respectively. More than half of the population (59.30%) in this study exceeded the safe limit of 1 ppm total hair mercury concentration, as recommended by the USEPA. Male participants from Yan had the greatest number exceeding that limit (15%).

3.2. Frequency of fish consumption

Respondents were asked about their fish eating habit, the frequency of fish meal and the amount of fish consumed. The amount of fish eaten per day is estimated from the number of fish meal multiplied by the weight of fish. An Indian mackerel measuring 19.5 cm by 5.0 cm, weighing 70 g, was shown to guide them to estimate the weight of fish consumed. Fig. 1 shows the difference between frequency in fish consumption between male and female participants. There was no significance difference in fish consumption between gender (*p= 0.053*). When they were grouped according to urban and rural areas, rural participants (Yan and Bachok) had more frequent fish meal compared to the urban participants (Alor Setar and Kota Bharu) (*p= 0.001*) (Fig. 2).

3.3. Amount of fish consumed

The amount of fish consumed ranged between 1.7 to 999.0 g/person/day. The average fish consumption was 180.19±11.34 g/day/person with a median of 140 g/person/day. Overall, men ate more fish compared to

Table 3. THg in hair by gender for all study locations

	THg (ppm)							
	Yan		Alor Setar		Bachok		Kota Bharu	
Statistic	Female (n=17)	Male (n=33)	Female (n=28)	Male (n=21)	Female (n=25)	Male (n=26)	Female (n=30)	Male (n=19)
Mean	0.98	1.92	1.16	1.10	1.50	1.33	1.14	1.35
Max	1.98	5.34	2.79	2.51	5.52	3.12	3.06	4.17
Min	0.53	0.63	0.34	0.46	0.48	0.63	0.43	0.04
Std deviation	0.33	1.06	0.55	0.50	1.11	0.69	0.71	0.90
Upper conf. limit	1.14	2.28	1.37	1.32	1.93	1.60	1.40	1.75
Lower conf. limit	0.82	1.56	0.96	0.88	1.07	1.06	0.58	0.95
Number of participants (%) exceeding1 ppm	7 (3.52)	30 (15.08)	15 (7.54)	11 (5.53)	18 (9.05)	14 (7.04)	13 (6.53)	10 (5.03)

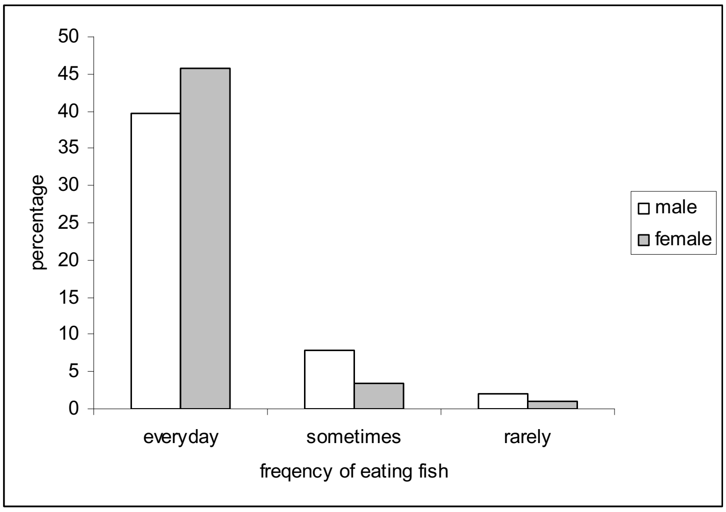


Figure 1. Frequency of fish consumption between male and female participants

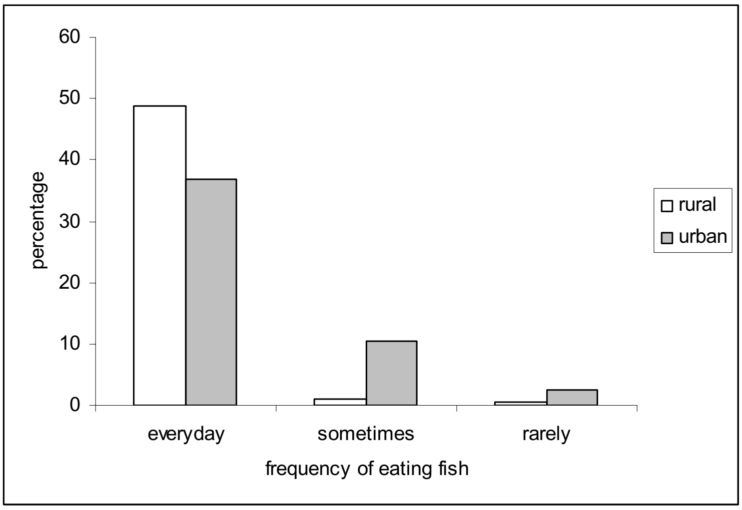


Figure 2. Frequency of fish consumption between rural and urban participants

women (*p=0.0001*). On average, men ate 195.38 g/day (95% CI=232.61; 269.85) and women ate 108.5 g/day (95% CI=130.54; 152.58). The urban residents consumed less fish compared to the rural residents (*p=0.0001*). The Yan and Bachok (rural) communities consumed an average of 331.21 g/day and 179.51 g/day while for Alor Setar and Kota Bharu (urban) communities the consumption were 110.88 g/day and 96.39 g/day, respectively (Fig. 3).

The participants were also asked to list the types of fish they usually eat. Fig. 4 shows the type of fish consumed at the different locations. Most of the participants consumed non-carnivorous, medium-sized (below 30 cm) marine fish. The most popular non carnivorous fish consumed was Indian mackerel (*Restrelliger sp*.). Among the carnivorous fishes consumed are Barred Spanish mackerel (*Scomberomorus sp*), Frigate tuna (Eastern little tuna) (*Auxis thazard*) and Red snapper (*Lutjanus sanguineus*). Further breakdown of fish consumption pattern based on locations are as follows. The top four most popular fish consumed by Kota Bharu residents were: Indian mackerel, Frigate tuna (Eastern little tuna), Barred Spanish mackerel, and Selar scad (*Selar sp*.). Bachok residents ate Indian Mackerel, Japanese scad (*Decapterus maruadsi*), Selar scad and Frigate tuna (Eastern little tuna). For Alor Setar residents, Indian mackerel, anchovy (*Stolephorus sp.),* Pomfret (*Pampus sp.*), and red snapper were eaten. Yan residents ate Indian mackerel, Barred Spanish mackerel, Pomfret, and Fringescale sardinella (*Sardinella fimbriata*). In general, the Indian mackerel topped the list for small-size non carnivorous fish (below 30 cm) in all areas. The big-size (above 30 cm) carnivorous fish (Barred Spanish mackerel) and the red snapper were popular among the west coast residents. However, residents in the east coast preferred Frigate tuna (big-size) and Selar scad (small-size).

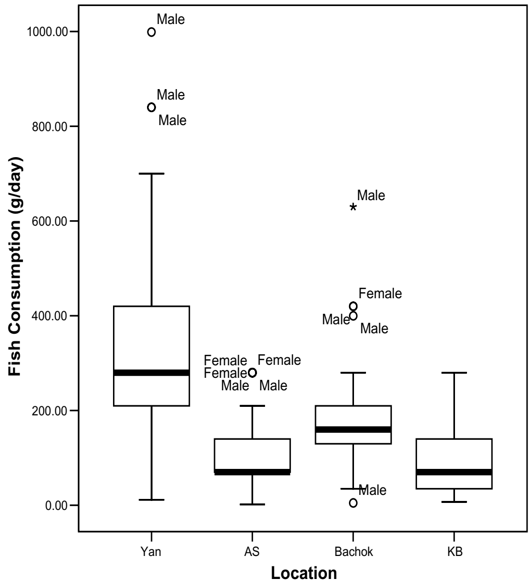


Figure 3. Comparison of amount of fish consumed by location and gender

3.4. Relationship between mercury exposure factors and THg concentration

The relationship between several exposure factors with THg is investigated in this work. In this work, seven factors were considered; gender, location of study, amount fish, age, body mass index (BMI), number of amalgam fillings and use of whitening cream. Statistical analysis showed that all factors were significantly correlated with THg. The amount of fish eaten was positively correlated with THg where the Spearman correlation coefficient (r_s) was 0.4199; (p=0.0001). Age was positively correlated with THg (r_s=0.4588; *p=0.0001*), followed by use of whitening cream (r_s=0.2410; *p=0.006*), BMI (r_s = 0.2034; *p=0.0041*), location of study (r_s=0.1818; *p=0.01*), and gender (0.1637; *p=0.0241*). However, we found negative correlation between the numbers of filling with THg (r_s=-0.2485; *p=0.004*). One possible explanation is that we observed that participants who lived in the rural areas tend to have fewer teeth. Figs. 5-8 illustrate scatter plots showing the relationship between THg and amount of fish, age, BMI and number of amalgam fillings. Fig. 9 illustrates the distribution of THg concentrations based on the frequency of fish consumed: Group 1= eat fish everyday; Group 2= sometimes (at least once a week); and Group 3= rarely (once a month). The group that reported eating fish everyday shows a pattern of increasing THg concentration compared to other groups. Fig. 10 illustrates the distribution of THg concentrations based on the amount of fish consumed. Fish consumption is categorized into the following groups: Group 1=< 100 gram/day; 2=>100-300 gram/day; 3=>300 gram/day. The group that reported eating more fish (>100 gram/day) shows a pattern of increasing THg concentration. The Kruskal-Wallis tests confirmed that there is a significant relationship between THg concentrations and frequency of fish consumed (*p<0.0001*); and THg concentrations and the amount of fish of fish consumed (*p<0.0001*).

4. Discussion

In this study, two female participants were excluded from the statistical analysis because their THg content was very high. Further investigation on these individuals revealed that inorganic mercury (IHg) contributed to the high THg, indicating that the source of mercury exposure is not through dietary intake. Use of cosmetics (whitening cream) and dental amalgams are possible sources of IHg. More than half of the population (59.30%) in this study exceeded the safe limit of 1 ppm total hair mercury concentration, as recommended by the USEPA. When we examined the female population, only 26.60% of the whole study group was above 1 ppm. Detailed breakdown of the surveyed female participants who were above 1 ppm revealed this information: Yan: 41. 18% (7/17) (range 0.53-1.98); Alor Setar: 53.57 % (15/28) (range 0.34 - 2.79), Bachok: 72% (18/25) (range 0.48 - 5.52); and Kota Bharu: 43.37% (13/30) (range 0.43 - 3.06). Only one individual exceeded the 5 ppm limit, a level associated with 5% risk of neurological lesions (WHO, 1990). None of them exceeded the 10 ppm level, which is associated with fetal toxicity. The use of this EPA's RfD value as a threshold of safe consumption for women is debatable for populations living in high fish

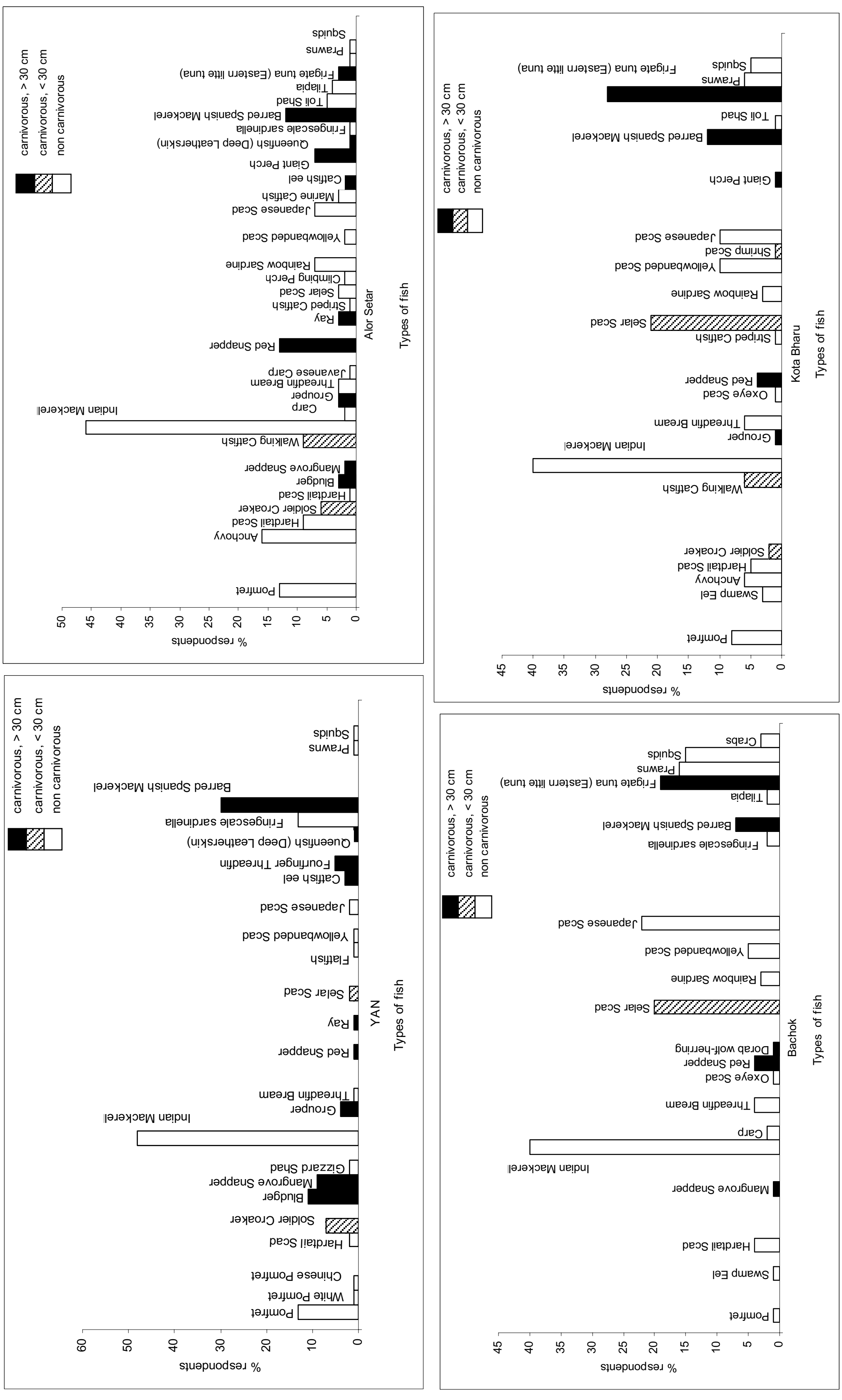


Figure 4. Types of fish consumed according to the location of study

3.4. *Relationship between mercury exposure factors and THg concentration*

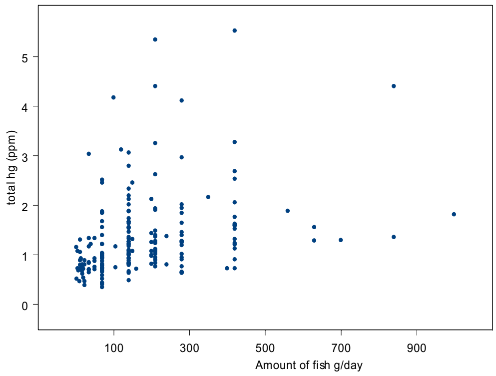


Figure 5. Scatter plots between amounts of THg with the amount of fish

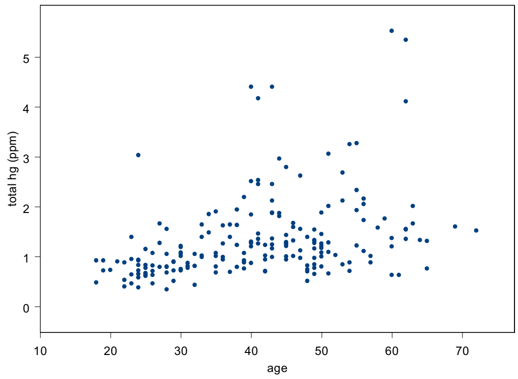


Figure 6. Scatter plots between amounts of THg with age.

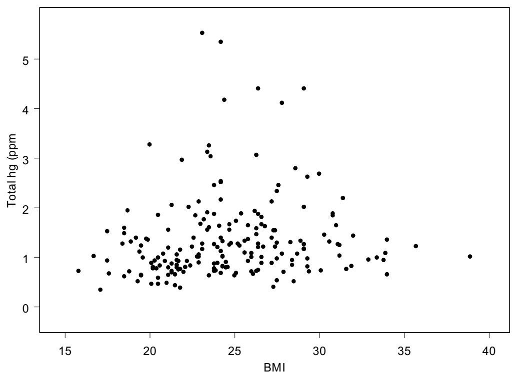


Figure 7. Scatter plots between amounts of THg with body mass index (BMI)

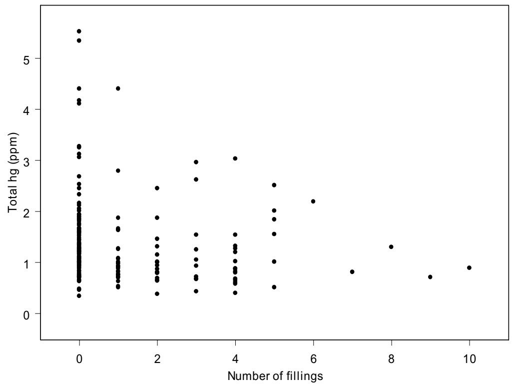
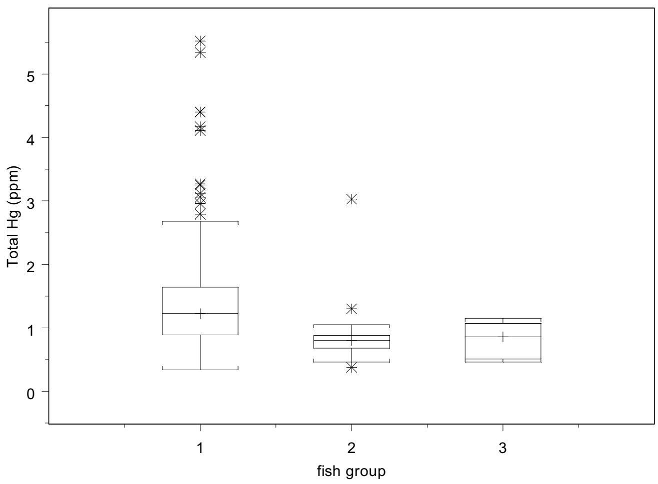
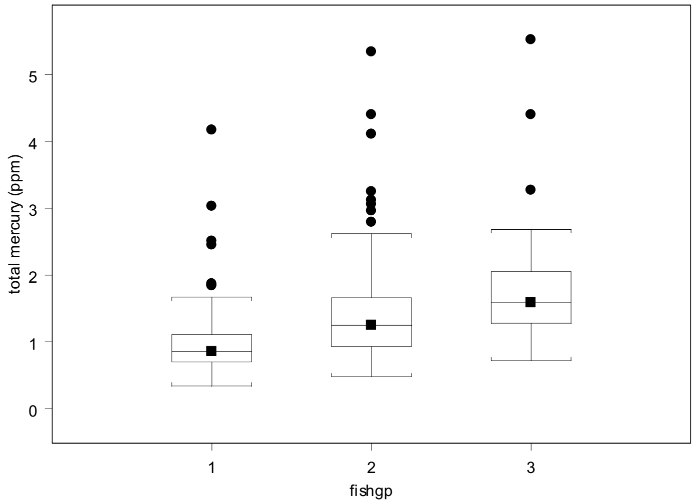


Figure 8. Scatter plots between amounts of THg with the number of amalgam fillings.



Group: 1= eat fish everyday; 2= sometimes (at least once a week); 3= rarely (once a month)

Figure 9. Distribution of hair mercury based on frequency of fish consumed.



Group: 1= < 100 g/d; 2= 100-300 g/d; 3= >300 g/d

Figure 10. Distribution of hair mercury based on the amount of fish consumed.

eating countries. Hair collected from 1666 Japanese women from five districts were estimated to be 1.42 µg/g (geometric mean), indicating that the level in general population might be higher (Yasutake *et al*., 2003). In Japan the safe exposure limit is 0.17 mg mercury/person/week which corresponds to hair mercury level to 5 µg/g (Yasutake *et al*., 2004).

This study found a significant relationship between gender, location of study, amount fish, age, body mass index (BMI), number of amalgam fillings, use of whitening cream and THg concentrations. Of the seven factors investigated, the amount of fish consumed and age showed the greatest correlation with THg. Findings for earlier studies showed that eating large carnivorous fish were implicated with elevated hair mercury levels (Malm *et al*., 1995; Dolbec *et al*., 2001; Yasutake *et al*., 2003). Knobeloch *et al*., (2007) reported that people who consumed sport- caught fish had mercury levels 1.5 times higher than those who ate commercial fish. Diez *et al*., (2008) reported positive correlation between fish consumption and THg for southern Italy urban population (r_s=0.536, $p<0.05$), suggesting a strong relationship between these two factors. In our study, most of the participants consumed mainly non-carnivorous, medium-sized marine fish. The use of fish consumption to estimate mercury exposure varies among studies. Majority of the studies expressed fish consumption as number of fish servings per week (month) or number of grams of fish per day. For our study, we used an actual fish, measuring 19.5 cm by 5.0 cm and 70 g in weight to guide the participants in determining the amount of fish consumed. According to the USEPA database, individual who consumes less than one serving of fish per month (100 g fish per month) are considered as no-fish-consumption population. Our study did not identify the non fish eaters.

The pattern of fish consumption observed in our study is influenced by gender and location of study. Although females have higher frequency fish meals, males consume more fish per meal. Again, this is reflective of the fact that most males who participated in this study were from rural (fishing) areas, where rural residents tend to consume more fish compared to the urban residents. Another factor that is related to the increasing mercury concentration is age. Again in this study, age is closely linked to location. Participants from rural areas (Yan and Bachok) were older, their mean age above 40 years, whereas participants from urban areas were in their mid 30's. Despite evidence of association between fish consumption and hair mercury concentration, we do not mean to suggest that people should refrain from eating fish. Our study did not sample mercury in fish; therefore, we were not able to calculate the actual amount of MeHg intake. However, previous studies have shown that increase in consumption of fish contaminated with mercury will result in increased mercury hair level (Santos *et al*., 2002; Weihe and Gradjean, 2006).

The public should be educated on risks and benefits of fish consumption. The benefit of eating small fish outweighs the risk of mercury exposure. According to Knobeloch *et al*. (2007), children and women of child bearing age should be limited to one-6 ounce meal or two 3 ounce meals per week. They suggested that fish such as northern pike and bass contain moderate mercury level, therefore should be limited to one meal per month. For big predatory fish, such as swordfish and shark, these should be avoided. Model simulation performed by Cohen *et al*. (2005)

showed that women of child bearing age can reduce MeHg exposure but maintain comparable n-3 PUFAs by changing their fish consumption pattern from consuming fish with high (>0.6 µg/g) or medium (0.14-0.6 µg/g) MeHg concentration to fish with low (<0.13 µg/g) MeHg.

In conclusion we found that 59.30% of the study population (26.60% female participants) has hair mercury levels exceeding the USEPA guideline of 1 ppm. Only one female participant exceeded the WHO recommended level of 5 ppm. The average fish consumption is 180.19 ± 11.34g/day/person with higher fish consumption in rural communities compared with the urban communities. The level of THg correlates with the following variables, arranged in decreasing Spearman correlation coefficient (r_s): amount of fish consumed per meal, age, use of whitening cream, BMI, number of amalgam fillings (negative correlation), location of study, and gender.

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