

PREDICT AND DESCRIBE RELATIONSHIPS BETWEEN THE LEVELS OF METHYLMERCURY IN HAIR SAMPLES OF KUALA LUMPUR RESIDENTS AND THEIR SOCIAL CHARACTERISTICS

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Abstract - A quasi-quadratic polynomial model is developed to predict and describe relationships between the level of methylmercury in hair samples of Kuala Lumpur residents and their social characteristics. The race, gender, age and fish diet factors appear to account for the ability to predict level of methylmercury. The model has a high explanatory power with coefficient of determination $R^2 = 88.4\%$. The results including interaction terms, showed a significant correlation at 1% level of significance. The results further revealed that the depicted rate of methylmercury level is heavily depending on the prevalence of the fish diet factor with an increasing rate.

INTRODUCTION

Mercury is a toxic element that released into the environment by human activities, such as combustion of fossil fuels and other industrial release. Once in the aquatic system microorganisms can convert inorganic mercury into methylmercury, the most toxic form of mercury which bioaccumulates in aquatic food webs including fish and shellfish. Therefore, fish and fish products are usually the main source of methylmercury in the diet¹. Because Malaysia is a peninsular state, fish is the main source of animal protein for the general population and covers about 80% of the protein requirement of the population. This puts the Malaysian population among the highest fish consumers in the world and exposure to methylmercury from fish consumption is, therefore, possible.

According to the World Health Organisation, the average people receive most of their mercury from fish and shellfish diets¹. Being fish constitutes the main source of protein for the great majority of Kuala Lumpur residents, it is important to study the influence of fish consumption on mercury intake by this population. The weekly consumption of fish meals by the Kuala Lumpur population ranged from zero to 14 times with the mean of 6 times per week.

Kuala Lumpur, which is located in a valley, is a city of undergoing industrialisation with a high population density. Fish diet is one of the major sources of protein for the residents of Kuala Lumpur community. All of these factors might result in a high mercury level for the residents. In this study, to assess the level of methylmercury exposure among the Kuala Lumpur population, hair samples were collected from donors living in different area of the city. Methylmercury was separated from hair samples prior determination by neutron activation analysis. The conceptual of quasi-quadratic polynomial modelling approach was developed and used to predict the level of methylmercury in the hair samples of the Kuala Lumpur local inhabitants. The independent variables included in the model were race, age, gender and fish diet.

MATERIALS AND METHODS

Sample collection

Kuala Lumpur, the capital of Malaysia, is the most populous, urbanised and industrialised region in the country. Kuala Lumpur is a plural society made up of three main ethnic groups: Malay, Chinese and Indian.

The sample of the study was composed solely of

randomly picked respondents of Kuala Lumpur residents. A stratified sampling method is applied by dividing the area into three strata. The number of the sample was 400 consisting of 46.25% Malay, 31.25% Chinese and 22.5% Indian.

Preparation of hair samples

Hair samples were collected from donors by single cutting from the occipital region with a pair of clean stainless steel scissors in accordance with the IAEA protocols. During collection of the hair samples, each individual was asked to complete a questionnaire detailing name, gender, age, occupation and dietary habits. The hair samples were cut to lengths of about 2-5 mm. The hair samples were then washed according to the standard procedure recommended by the IAEA: wash hair in acetone, thrice in water and once more in acetone. The samples were then dried overnight in an oven at 60°C.

Determination of total mercury and methylmercury

The prepared hair samples were analysed by neutron activation analysis for total mercury and methylmercury determination. In order to separate methylmercury from hair samples prior to neutron activation analysis, a simple technique was developed. The procedure is based on digestion of the hair samples in NaOH, extraction of methylmercury into toluene followed by back extraction in filter paper impregnated with cysteine. The applied method was described in details in our previous work 2.

Data analysis

Data analysis, ANOVA and ridge regression were performed using the STATISTICA software. A quadratic polynomial model containing nine coefficients, including interaction terms, was assumed to provide information on the relationships among the factors that may account for the inhabitants' methylmercury concentrations:

$$Y = \beta_0 + \sum_{i=1}^3 \beta_i D_i + \sum_{i=1}^2 \beta_i X_i + \beta_{ii} X_i^2 + \beta_{ij} X_i X_j + \beta_{ij} D_3 X_i + \mu$$

where Y denotes the level of methylmercury concentration in hair measured in mgkg⁻¹. β_0 is the constant coefficient (intercept), β_i the linear coefficient, β_{ii} the quadratic coefficient and β_{ij} the second-order interaction coefficient.

D_i are dummy variables representing the race and gender factors. Two dummies are used to represent Chinese (D_1) and Indian (D_2) races. Malay race is control group. D_3 is dummy variable for gender (1=female, 0=male). X_i refers to the age of the respondent

measured in years (X_1) and to the amount of fish diet consumed per respondent per week measured in kg (X_2). X_2^2 is a squared term of consumed amount of fish diet per respondent. $X_1 X_2$ refers to an interaction between age and consumed amount of fish diet. $D_3 X_2$ is an interaction between gender and consumed amount of fish diet. The error term μ is assumed to capture unobserved respondent specific characteristics that are not accounted for in the intercept terms and which might create a differential impact on methylmercury exposure.

RESULTS AND DISCUSSION

Sample social characteristics

The empirical results of the survey revealed that 47% and 53% of the randomly interviewed respondents within the two types of gender have an age of 2 to 80 years old. This constitutes 46%, 31% and 23% of the Malay, Chinese and Indian races, respectively. The mean (\pm standard deviation) age of the whole population was 32.80 \pm 15.95 years and those for ethnic Malays, Chinese & Indians were 33.28 \pm 16.31, 33.66 \pm 16.26 and 30.61 \pm 14.68, respectively. Table 1 summarizes the characteristics of the sample of the study.

Distribution of hair methylmercury

The overall average level of methylmercury in hair samples of the inhabitants was 1.13 mg kg⁻¹ in the range of 0.00-4.465 mg kg⁻¹ and form 31.15 % of total mercury in the range of 0-75.81%. For the male, the average level of methylmercury concentrations was 1.03 mg kg⁻¹ (ranged from 0.00 to 3.87 mg kg⁻¹) and for the female was 1.25 mg kg⁻¹ (ranged from 0.00 to 4.65 mg kg⁻¹).

Table 2 shows the ethnic distribution of hair total mercury and methylmercury levels. The highest level was found in the Chinese race followed by the Indian race then the lowest level was found in the Malay race.

Results of the analysis of ridge regression between the levels of methylmercury in hair and the various examined contributing variables are presented in the Table 3. Multicollinearity problem among the independent variables was observed. Therefore, ridge regression analysis is used to obtain more stable estimates that cannot be obtained via ordinary least square methods. The evidences of heteroscedasticity & autocorrelation problems were inconclusive.

The high explanatory coefficient of determination indicating that 88% of the variability in methylmercury concentration levels among the

inhabitants hair of Kuala Lumpur is explained by the race, gender, age and fish diet factors. Further, the ANOVA F-value of 380.46 indicates the overall significance of the developed model at 1% (Table 4). This means the rejection of null hypothesis that the regression coefficients $\beta_1 = \beta_2 = \beta_3 = \dots = \beta = 0$ and the acceptance of the alternative hypothesis that at least one factor coefficient is not equal to zero. Moreover, it indicates that such association between the mercury concentration levels and the contributing factors under consideration could not be of random origin.

All terms regardless of their significance were included in the following quasi second-order polynomial equation :

Table 1. Distribution of ethnic group according to age and gender

Ethnic group	N	%	Age (years)	Male %	Female %
Malay	185	46.25	2 - 80	45.90	54.10
Chinese	125	31.25	4 - 75	49.60	50.40
Indian	90	22.50	3 - 70	53.33	46.67
Total	400	100 %	2 - 80	47.25	52.75

Table 2. Distribution of total mercury and methylmercury levels within the ethnic group

Ethnic group	T-Hg		Me-Hg		% Me-Hg	
	Range	Median	Range	Median	Range	Median
Malay	0.83-15.30	2.75	0.0-4.00	0.75	0.0 - 57.85	28.16
Indian	0.59-18.73	3.81	0.0-4.02	1.31	0.0 - 70.18	30.67
Chinese	0.91-15.69	4.50	0.0-4.65	1.52	0.0 - 75.81	33.96
All	0.59-18.73	3.38	0.0-4.65	1.13	0.0 - 75.81	31.15

Table 3. Ridge Regression results of a quasi-quadratic polynomial model for methylmercury exposure

Term	Coefficient	Standard error	T-value	P-value
Constant	-0.159			
Main effects				
Chinese race	0.359**	0.034	10.504	0.000
Indian race	0.287**	0.036	7.876	0.000
Gender	-0.020	0.064	-0.309	0.757
Age	0.001	0.002	0.383	0.702
Fish diet	0.742**	0.166	4.470	0.000
Squared effect				
Fish diet* Fish diet	1.043**	0.105	9.969	0.000
Interaction effects				
Gender*Fish diet	0.343**	0.094	3.651	0.000
Age*Fish diet	0.007*	0.003	1.978	0.049

Lambda = 0.023; R = 94.1%; R2 = 88.6%; Adjusted R2 = 88.4%; Predicted R2 = 88.02%; Standard error of the response = 0.285 ; DF = 8 ; *Significant at 5%; ** Significant at 1%.

$$Y = -0.159 + 0.359 \text{ Chinese race} + 0.287 \text{ Indian race} - 0.020 \text{ Gender} + 0.001 \text{ Age} + 0.742 \text{ Fishdiet} + 1.043 \text{ Fishdiet} * \text{Fishdiet} + 0.343 \text{ Gender} * \text{Fishdiet} + 0.007 \text{ Age} * \text{Fishdiet}$$

According to our modelled findings (Table 3), there is a significant contribution of Chinese (0.36) and Indian (0.29) races to the level of methylmercury among residents of Kuala Lumpur community. These high levels of Chinese and Indian races contribution could be attributed to the difference in fish diet consumption frequency between the three races. Our results revealed that Chinese and Indian groups consume more fish diet than Malay group. A total of 79% of Chinese group and 63% of the Indian group consume fish in range of 6-14 times/week, while 51% of the Malay group consume fish in the same range.

The regression coefficient of gender factor showed insignificant and negative contribution to the level of methylmercury of the Kuala Lumpur population (Table 3).

This result agrees with those reported by Kyle & Ghani8 and Barbosa *et al.* 2001. However, the interaction between gender and fish diet showed

positive and significant contribution to the level of methylmercury among the studied population. This finding indicates that female consumes more fish than male. It is inconsistent with Akagi *et al.* 1995; Malam *et al.* 1995, who found that, males consume more fish than females and as a consequence males had higher level of methylmercury than females. However, the descriptive measures of the present study revealed that 30% of the Kuala Lumpur population consumed fish diet. An average of 10 times per week of consumed fish diet is figured out in 63% of this population (29% male, 34% female).

The regression coefficient of age in Table 3 showed a lowest and insignificant effect on methylmercury levels. Likewise, the interaction between age & fish diet was positively and significantly contributed to the level of methylmercury. This result revealed that, the older people of Kuala Lumpur community tended to consume more fish diet than younger.

Table 4. Analysis of variance for the ridge regression model of methylmercury exposure

Source of variation	Sum of squares	Degrees of freedom	Mean squares	F-value	P-value
Regression	247.710	8	30.964	380.456**	0.000
Residual	31.822	391			
Total	279.531				

** Significant at 1%.

As expected, the amount of fish diet consumed per week per resident of the Kuala Lumpur inhabitants correlated positively and significantly to the level of methylmercury concentration. This correlation is relevant to our understanding of inhabitants' methylmercury exposure and it is therefore useful for predicating the methylmercury exposure among the target population. Such results are in agreement with those reported elsewhere for fish diet consumer populations 3-9.

The fish diet which is the core factor among the others in this study and as a main effect resulted in a largest contribution to the concentration levels of methylmercury. This might give an indication of polluted environment in fishery producing farms. Also the regression coefficient factor with the largest effect was the squared term of fish diet. This indicates that fish diet contribution is distinguishable from other factors and could be evaluated as a curvature (quadratic). This evaluation showed that the depicted rate of methylmercury level is heavily depending on the prevalence of the fish diet factor, and it is increasing on an increasing rate of 1.04 units (Table 3). That is, the relationship between level of methylmercury and consumed amounts of fish diet follow a curve line, rather than a straight line.

The interaction effects resulted in the fact that both gender and age factors depend on fish diet factor to contribute to the variability of methylmercury levels among the residents of Kuala Lumpur.

The constant $\lambda=0.023$, ridge regression, is added to the diagonal of the correlation matrix, which is then re-standardized so that all diagonal elements are equal to 1.0 and the off-diagonal elements are divided by the constant. In other words, it decreases the correlation coefficients so that more stable estimates can be estimated.

The correlation coefficient R is generally used to provide correlation measures for the estimation of the regression model. The closer the value of R to unity, the better is the correlation between the observed and the predicted values. The value of R for the level of the methylmercury was 0.941. This value of R indicates a good agreement between the observed and predicted values of the level of the methylmercury. On the other hand, the predicted R^2 88.02%

measures the amount of variation in new data explained by the developed model.

CONCLUSION

We have developed a quasi-quadratic polynomial model for the relationships between the concentration levels of methylmercury exposure among the local inhabitants of Kuala Lumpur and some of their social factors. Our results indicate that, the proposed model was reasonable for evaluating the level of methylmercury among the Kuala Lumpur residents. Further, the relationship between the level of methylmercury in hair of the residents and these factors is useful when presented in terms of a quasi second-order polynomial regression model. According to the present work, fish consumption is highly and significantly contributed to the level of methylmercury in hair samples of Kuala Lumpur population. Likewise, Chinese and Indian races contributed significant to the level of methylmercury. As an additional advantage these significant results of this developed model would leave a platform to others to formulate appropriate models for the purpose of building explicit, and theoretically interesting comparisons into our model.

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