Risk Assessment Studies Report No. 13

Chemical Hazard Evaluation

OF SECONDARY SCHOOL STUDENTS

(Follow-up Report)

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Food and Environmental Hygiene Department

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Abstract

In 2002, the Food and Environmental Hygiene Department conducted a study on "Dietary Exposure to Heavy Metals of Secondary School Students" and assessed whether there is any risk to the health of secondary school students resulting from the dietary exposures to heavy metals. In view of the limitations in the previous study and the availability of methylmercury testing locally, a follow-up study on dietary exposure to total mercury and methylmercury was conducted to re-examine the issue with a view to obtaining more precise estimates of dietary exposure to mercury.

Dietary exposures to total mercury and methylmercury were estimated by using the local food consumption data obtained in secondary school students and the concentrations of total mercury and methylmercury in food samples taken from the local market specifically for the study. More sophisticated laboratory analysis was employed with lower limit of detection.

The results of the study showed that the dietary exposures to total mercury and methylmercury were all within their respective Provisional Tolerable Weekly Intakes (PTWIs) for both average and high consumers. The dietary exposures to total mercury and methylmercury for average secondary school students were 0.92 and 0.35 μ g/kg bw/week respectively and that for high consumers were 2.33 and 0.87 μ g/kg bw/week respectively. It can be concluded that both the average consumers and high consumers among secondary school students would be unlikely to experience major toxicological effects of total mercury and methylmercury.

The results also showed that the food group "fish" was identified as the main dietary source of total mercury and methylmercury, and the large predatory fish, such as swordfish and tuna, had the highest concentration of total mercury and methylmercury.

Dietary Exposure to Mercury of

Secondary School Students

(Follow-up Report)

OBJECTIVE

This is a follow-up study aiming to (i) estimate more precisely the dietary exposures to total mercury and methylmercury by the secondary school students in Hong Kong and (ii) assess the health effects of total mercury and methylmercury resulting from these exposures.

BACKGROUND

The Previous Study

2. The Food and Environmental Hygiene Department (FEHD) conducted a study on "Dietary Exposure to Heavy Metals of Secondary School Students" ¹ in 2002. The study aimed to determine the dietary exposure to heavy metals of secondary school students in Hong Kong so as to assess whether there are any risks to their health.

Three heavy metals, namely arsenic, cadmium and mercury, were chosen for the study.

- 3. The dietary exposure of secondary school students to the heavy metals were estimated in the previous study using two sets of data. The levels of heavy metals were extracted from the database of the Food Surveillance Programme of FEHD and the food consumption data was derived from the Food Consumption Survey² conducted by FEHD in late 2000.
- 4. The previous study estimated that the dietary exposure to total mercury for an average secondary school and high consumer were 2.98 and 6.41 μ g/kg bw/week respectively. The exposure estimate for an average secondary school fell within the Provisional Tolerable Weekly Intake (PTWI) of 5 μ g/kg bw/week while that for high consumer exceeded the PTWI. "Cereal and cereal products" and "vegetables" were found to be important dietary sources of total mercury and this finding was not consistent with those reported in the literature.
- 5. It was noted that there were certain limitations in the previous study. In the Food Surveillance Programme, which is mainly for enforcement purposes, analyses for mercury were determined in form of total mercury rather than methylmercury. The limit of detection (LOD) was 30µg/kg, which was considered high if used for research studies.
- 6. A value of 1/2 LOD was assigned to samples with

levels below LOD as a conservative approach and the majority of samples for the two food groups "cereal and cereal products" and "vegetables" have mercury levels below LOD. The relatively high LOD employed by the Food Surveillance Programme and the high amount of consumption for these two food groups might have exaggerated the exposures from them.

With the availability of methylmercury testing and the concern over its exposure and the limitations in the previous study, FEHD considered it necessary to re-examine the issue with a view to obtaining a more precise estimate of dietary exposure to mercury. More sophisticated methods of analysis on both total mercury and methylmercury would be developed with a lower detection limit. Food samples would be collected specifically for the study.

Mercury

8. Mercury (Hg) exists naturally in abundance in the environment. It enters the environment by both natural and human means. Mercury exists in three forms, namely metallic, inorganic and organic mercury. The forms can be altered under certain conditions. Mercury can be oxidized to inorganic bivalent mercury with the presence of organic matters in waters and can be converted back to metallic mercury in certain industrial effluent. The inorganic mercury can also be converted to methylated form by microorganisms especially in aquatic

systems. Methylmercury bio-accumulates in the food chain and thus can result in high concentrations in large predatory fish.³

Sources of Exposure

- 9. Contamination of food from both natural and human sources, dental amalgam and occupational exposure in agriculture and manufacturing sectors are possible routes of exposure to the chemicals. The dietary intake is the most dominant source of exposure to mercury.⁴
- 10. In most foodstuffs, mercury is largely in the inorganic form. However, fish and other seafood products are the main source of methylmercury, of which large predatory species such as swordfish and tuna tend to accumulate relatively higher levels. Methylmercury bio-accumulates as it moves up the food chain, increasing in concentration at the same time. ⁴

Toxicological Effect

- 11. Mercury and its compounds have no known physiological functions in animals. Their presence in human is considered as undesirable and may be hazardous to health.⁵
- 12. Organic mercury compounds are more harmful than inorganic mercury. Methylmercury is the most common form of organic mercury and is regarded as highly toxic.

- 13. Methylmercury can cause adverse effect to the nervous system, especially the developing brain. Methylmercury passes more readily through the placenta than the other mercury compounds. Foetus exposed to methylmercury have been found to be born mentally retarded and with symptoms similar to those of cerebral palsy. Pregnant mothers who received low level of methylmercury by normal adult standard may give birth to children with serious cerebral palsy. Hence, unborn foetus, infant and young children are more sensitive to such toxic effects. ⁴
- 14. Acute toxicity is often a result of occupational exposure, and that from dietary exposure is rare. Acute effects include increased occurrence of lymphocytic aneuploidy, discolouration of the front surface of lens of the eyes, insomnia, tremors and hyperexcitability.
- 15. There is a long latent period before early symptoms of methylmercury poisoning including paraesthesia, malaise and blurred vision emerge. Constriction of the visual field, deafness, dysarthria and ataxia may develop at a later stage. The patient may partly recover from the symptoms in a less severe case or may fall into a coma as a result in a severe case. Damages to the central nervous system are highly localized and affect mostly sensation, vision and hearing. ⁴
- 16. The Joint FAO/WHO Expert Committee on Food Additives (JECFA) has established the PTWIs of $5\,\mu\text{g}/\text{kg}$ bw/week for

total mercury and 3.3 μ g/ kg bw/week for methylmercury respectively. Since foetus and infants may be at greater risk of toxic effects, JECFA in July 2003 reduced the PTWI for methylmercury to 1.6 μ g/kg bw/week and this level is considered sufficient to protect the developing foetus, the most sensitive subgroup of the population.^{6, 7}

17. PTWI is an estimate of the amount of a contaminant that can be ingested over a lifetime without appreciable risk.

SCOPE OF STUDY

18. To estimate the dietary exposures to total mercury and methylmercury, this study covered six major food groups, namely (i) cereals and cereal products, (ii) vegetables, (iii) meat, poultry and their products, (iv) fish, (v) seafood other than fish, and (vi) milk and dairy products. The other food sources, such as beverages, were not covered in this study as they were generally not regarded as significant sources of mercury.

METHODOLOGY

Food Consumption Data

As in the previous study, the food consumption data in this report were extracted from the Food Consumption Survey conducted on local secondary school students in 2000 by the Department. Readers may refer to report of previous study¹ and the Food Consumption Survey report for details.²

Sampling Plan for Food Samples

Food samples specific for the present study were taken from the local market according to the six food groups as mentioned in para. 18. The food items in each food group were selected so as to match those in the Food Consumption Survey.

Laboratory Analysis

Laboratory (FRL) of FEHD and the analysis was conducted mainly in form of "composite sample". Samples of the same food types were mixed and homogenized. For total mercury analysis, the food samples were digested with concentrated nitric acid and hydrogen peroxide, and the mercury content was determined by cold vapour atomic absorption spectrometry with amalgamation as a pre-concentration step. For

methylmercury analysis, methylmercury in the food sample was extracted by hydrochloric acid and derivatized with sodium tetraphenylborate, and was analyzed by gas chromatography equipped with atomic emission detector. The LOD for total mercury and methylmercury for this study were 3 μ g/kg and 1 μ g/kg (calculated as mercury) respectively.

- When the analytical value was below the LOD, the true value could be anywhere between zero and the LOD. The treatment for these results was particularly important when a large percentage of the analytical results of a particular food group were below LOD.
- 23. While it may not be appropriate to assume a zero concentration for all the samples with analytical values below LOD, assigning the non-detects the value of LOD would, however, grossly overestimate the dietary intake. A value of 1/2-LOD was assigned to all results below LOD in this study. Since the levels of contaminants in food, including mercury, usually follows a log-normal distribution, assigning a value of 1/2 LOD to all non-detected levels is considered as a conservative approach, especially for food groups in which the majority of food items have levels below the LOD.
- 24. For non seafood samples, only those samples with analytical values of total mercury above LOD were further subject to methylmercury analysis since mercury in non seafood items is largely in the inorganic form. For these samples, a value of 1/2-LOD for methylmercury was also assigned as a conservative approach.

Dietary Exposures to Total Mercury and Methylmercury

- Dietary exposure from individual food item was obtained by combining the consumption data and the median concentration of individual food items. Total exposure for each student was obtained by summing exposures from all food items. The mean of the exposures of all the students was used to represent the average dietary exposures. The 95th percentile of the exposure level was used to represent the exposure for high consumers.
- 26. The daily dietary exposure was multiplied by seven to obtain a weekly exposure level of an average secondary school student.
- 27. The estimated weekly exposure level was then compared with the PTWI as established by JECFA.

RESULTS

Concentration of Total Mercury and Methylmercury

A total of 347 food samples were taken and combined into 115 composite samples for analysis. The median concentrations for total mercury and methylmercury for each food group are given in Table 1.

Table 1: Median Concentrations for Total Mercury and Methylmercury in Six Food Groups

Food group	Total Mercury		Methylmercury	
	% of samples Median		% of samples	Median
	below LOD	concentration (mg/kg)	below LOD/ not analyzed	concentration (mg/kg)
Cereal and cereal products	100.0	1.5 *	100.0	0.5 *
Vegetables	91.7	1.5 *	100.0	0.5 *
Meat, poultry and their products	94.4	1.5 *	100.0	0.5 *
Fish	0	58	0	19
Seafood other than fish	0	20	0	6
Milk and dairy products	100.0	1.5 *	100.0	0.5 *

(* a value of 1/2 LOD was assigned)

29. Total mercury and methylmercury were detected in all the samples from the two food groups "fish" and "seafood other than fish". For the remaining food groups, only a few samples were found to contain low levels of total mercury but none of them was found to contain methylmercury. Distribution curves of total mercury and methylmercury concentrations in the two food groups "fish" and "seafood other than fish" are presented in Annex I.

Dietary Exposures to Total Mercury and Methylmercury

Average Secondary School Students

30. The estimated dietary exposures to total mercury and methylmercury for average secondary school students were obtained from the mean exposure level. The dietary exposures for average secondary school students were 0.92 and 0.35 μ g/kg bw/week for total mercury and methylmercury respectively. The results are shown in Table 2.

Table 2: Estimated Dietary Exposure to Total Mercury and Methylmercury for Average Secondary School Students

Food Group	Dietary Exposure in mg/kg bw/week (% contribution)			
	Total N	Mercury	Methy	lmercury
Cereals and cereal products	0.10	(11%)	0.03	(10%)
Vegetables	0.07	(8%)	0.02	(6%)
Meat, poultry and their products	0.04	(4%)	0.01	(4%)
Fish	0.54	(59%)	0.21	(59%)
Seafood other than fish	0.13	(14%)	0.06	(18%)
Milk and dairy products	0.03	(3%)	0.01	(3%)
Total	0.92	(100%)*	0.35	(100%)*

^{(*} Figures may not add up to total due to rounding)

High Consumers

31. Further analyses were undertaken to estimate the risk that high consumers might be exposed to. The 95th percentile exposure

level of the secondary school students was used to represent the dietary exposure to total mercury and methylmercury for high consumers. The dietary exposures for high consumers were 2.33 and 0.87 μ g/kg bw/week for total mercury and methylmercury respectively. (Table 3)

Table 3: Comparison among PTWIs Established by JECFA,
Dietary Exposures for Average Secondary School
Students and High Consumers for Total Mercury and
Methylmercury

	JECFA PTWI (mg/kg bw/week)	Exposure (mg/kg bw/week) (% of PTWI)	
		Average Secondary School Students	High Consumers
Total Mercury	5	0.92 (18%)	2.33 (47%)
Methylmercury	1.6	0.35 (22%)	0.87 (54%)

DISCUSSION

- 32. For an average secondary school student, dietary exposures to total mercury and methylmercury were 0.92 and 0.35 μ g/kg bw/week respectively. The proportions of their respective PTWIs contributed by these exposures were 18% for total mercury and 22% for methylmercury. The exposures were well below the respective PTWIs established by JECFA for both total mercury and methylmercury.
- 33. For high consumers, the dietary exposures to total mercury and methylmercury were 2.33 μg/kg bw/week (47% of PTWI)

and $0.87~\mu g/kg$ bw/week 64% of PTWI). Exposures to total mercury and methylmercury for high consumers were also below their respective PTWIs.

- 34. Therefore, the dietary exposure estimates of our results suggested that secondary school students for both average and high consumers were unlikely to experience major undesirable health effects to total mercury and methylmercury.
- 35. In our findings, the main dietary source of total mercury and methylmercury was "fish" which contributed 59% of both total mercury and methylmercury exposures. This is followed by the food group "seafood other than fish" (14% of total mercury and 18% of methylmercury exposures respectively). The findings are consistent with those conducted in overseas dietary exposures studies such as those conducted in the US, UK and Australia. However, this differs from the results of our previous study which had overestimated the contribution of cereal and cereal products because of the limitation of the study and had therefore identified cereal and cereal products as the main contributor (34%) of dietary mercury.
- 36. The laboratory analysis used in the present study with a lower detection limit as compared with the previous study provided a more precise estimation of the dietary exposure pattern. In the previous study, the LOD for total mercury was 30 μ g/kg while in the present study, the LOD for total mercury was 3 μ g/kg and that for methylmercury was 1

μg/kg. Even with the low LOD, most of the results in the non-seafood food groups were still below LOD. Because of the approach to assign 1/2 LOD instead of assigning zero to levels below LOD, the exposure estimate from individual non-seafood food groups or food items would be directly proportional to the LOD.

- 37. Because of the relatively high amount of consumption, "cereal and cereal products" was still estimated to contribute 11% for total mercury and 10% for methylmercury exposures respectively.
- 38. The majority of non-seafood items have total mercury levels below LOD. Only those non-seafood samples with detectable levels of total mercury were further analyzed for methylmercury. For non-seafood samples with total mercury level below LOD, a value of 1/2 LOD for both total mercury and methylmercury was assigned to these samples. Since it is expected that mercury in food other than seafood is largely in the inorganic form⁸, this conservative approach would attribute to an overestimation of the methylmercury exposure from these food When a methylmercury level of zero is assigned for groups. non-seafood samples with non-detected total mercury, the dietary exposure to methylmercury for average and high consumers would become 0.27 and 0.76 µg/kg bw/week respectively and the contributions of food groups "fish" and "seafood other than fish" would become 76% and 23% respectively.
- 39. Among the six food groups, "fish" has the highest

levels of both total mercury and methylmercuy, and this is followed by "seafood other than fish". Of the food items analysed, large predatory fish such as swordfish and tuna had the highest level of mercury. This finding is similar those obtained in our previous study and in overseas studies. The percentages of total mercury being methylmercury in "fish" ranged from 18% to 96% with a median of 40% and that in "seafood other than fish" ranged from 13% to 95% with a median of 49%.

Estimates of dietary exposure to total mercury were compared with findings in overseas studies and presented in Table 4. However, direct comparison of the data has to be done with caution due to the differences in research methodology, food group categorization, methods of collection of consumption data, methods of contaminant analysis and methods of treating results below detection limits. The dietary exposure estimated in our study is comparable to exposure patterns obtained from overseas studies.

Table 4: A Comparison of Average Weekly Exposure of Total Mercury

Country	Average Weekly Dietary Exposure (mg/person/week)
USA	8.75
Netherlands	14
UK	21.7
Hong Kong	47.8 *
New Zealand	51
Japan	72
China	72.1
Australia	26-126
Spain	129

[#] Data contained in this table is extracted from the report on "Dietary Exposure to Heavy Metals of Secondary School Students" published by FEHD in 2002.

JECFA has established a PTWI of 1.6 μ g/kg bw/week for methylmercury for protection of the developing foetus, the most sensitive subgroup of the population. Since the data of consumption pattern for pregnant women is not available, the health risk of methylmercury resulting from dietary exposure of pregnant women can not be assessed. However, the 95th percentile exposure level obtained in our study (0.87 μ g/kg bw/week) is still below this PTWI.

Limitation

42. The methodology for collection of food consumption data may influence the accuracy of the estimates on dietary exposure. In the Food Consumption Survey, the food consumption pattern of secondary school students was collected using a food frequency

^{*} The exposure data in Hong Kong is calculated by multiplying the dietary exposure for average secondary school students and mean body weight (i.e. $0.92~\mu g/kg~bw/week~X~52.0~kg$).

questionnaire. Although the food frequency questionnaire was very comprehensive, it was not possible to cover every single food item, some of which might be relevant to mercury exposure. For example, the consumption pattern of individual seafood species is not known and thus the dietary exposure from seafood products was estimated from the consumption of food groups "fish" and "seafood other than fish".

CONCLUSIONS AND RECOMMENDATIONS

- The dietary exposures to total mercury and methylmercury for average secondary school students were 0.92 and 0.35 μ g/kg bw/week respectively and that for high consumers were 2.33 and 0.87 μ g/kg bw/week respectively. They are all within their respective PTWIs established by JECFA. It can be concluded that both the average and high consumers among secondary school students would be unlikely to experience major toxicological effects of total mercury and methylmercury.
- 44. The food group "fish" was identified as the main dietary source of total mercury and methylmercury. The large predatory fish such as swordfish and tuna had the highest concentration of mercury and they may be significant dietary sources of mercury especially when they are consumed in large amount.
- 45. A balanced diet is essential to avoid excessive

exposure to mercury from a small range of food items.

Susceptive groups such as children and pregnant women should be careful in the selection of food, in particular, they are advised not to consume excessive amount predatory fish such as swordfish and tuna. While fish is a good source of high-quality protein and low in saturated fat, and moderate consumption of fish is recommended.

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ANNEX I – Distribution Curves of Total Mercury and Methylmercury Concentrations in Fish and Seafood other than Fish







