Hair Mercury Levels of Residents in China, Indonesia, and Japan

QIUYANG FENG
YASUO SUZUKI
AKINORI HISASHIGE
Department of Hygiene
School of Medicine
Tokushima University
Kuramoto-cho, Tokushima, Japan

ABSTRACT. The authors used gold-amalgamation cold-vapor atomic absorption spectrometry and ECD-gas chromatography to analyze total mercury and methylmercury levels in hair samples obtained from 362 residents in Harbin, China; Medan, Indonesia; and Tokushima, Japan. In this study, the authors initially questioned whether mercury levels in hair differed among different study areas, and if there were differences, they questioned the contributing factors. In the three countries surveyed, total mercury and methylmercury levels in hair were lowest in residents of China and were highest in residents of Japan. In the district of Tokushima, Japan, total mercury and methylmercury levels were highest in the coastal district, followed by the middle district; the lowest levels occurred in the mountainous district. In Japan, an individual’s total mercury level correlated very closely with that person’s methylmercury level; in China and Indonesia, the correlation between these 2 parameters was low. No subjects in China or Indonesia had high levels of methylmercury in hair; this was true even if their total mercury levels were high. This finding suggests that the high total mercury levels observed in some residents of China and Indonesia reflected exposure to inorganic mercury. In Japan, mercury (especially methylmercury) levels in hair samples were quite high. Fish and shellfish, caught in seas uncontaminated by human activity, appeared to be major sources of the high levels of hair mercury in Japanese subjects.

DURING THE PAST TWO DECADES, there has been worldwide concern about the high toxicity and ubiquitous nature of environmental mercury contamination.1-8 Investigators have focused their concern mainly on the presence of elevated mercury levels in seafood, especially fish, which is a major human dietary source of mercury. Most of the mercury concentrates in the form of methylmercury in the edible tissue of fish; therefore, fish and fish products are usually the main source of methylmercury in the diet.9,10

People accumulate mercury in their bodies primarily via absorption of mercury from their diets (especially fish) and from air in their local environments.12 Fish consumption, country and place of residence, occupation, age, and sex all have some effect on hair mercury levels.13-18

The existence of mercury, especially methylmercury, in marine fish has been treated in an extensive review, as well as in proceedings of international conferences on mercury19; the results have prompted researchers to undertake urgently needed studies on mercury in the environment. In the present study, we examined hair mercury levels of 362 randomly selected residents (i.e., males aged 40–49 y) from Harbin, China (approximately 600 km from the coast); Medan, Indonesia (about 30 km from the coast); and different districts of Tokushima, Japan (contiguous to the Pacific Ocean). We sought to determine if hair mercury levels varied among study areas located different distances from the coast.

Investigators have used various specimens from organisms (e.g., blood, nail, urine, hair) to assess mercury levels in the human body.20-28 We chose to use hair in our study because sampling and storage of hair is easier than sampling and storage of other biological materials.29 In many studies, researchers have shown that, under certain conditions, hair can be a good indi-
Material and Method

Collection of samples. In this study, we used 362 hair samples obtained from male residents between the ages of 40 and 49 y. Of these samples, 64 were from Harbin, China (approximately 600 km from the coast); 55 were from Medan, Indonesia (about 30 km from the coast); and 243 were from the Tokushima Prefecture in Japan (contiguous to the Pacific Ocean). Of the 243 Tokushima samples, we obtained 39 from (a) a mountainous district farther from the coast than the other two districts (i.e., middle and coastal districts) of this prefecture; (b) 78 from a coastal district facing the Pacific Ocean; and (c) 126 from a middle district located between the mountainous district and coastal district. The results of the 1995 report from the Tokushima Prefecture environmental white paper indicated that the three districts of Tokushima did not have any contamination induced by human activity (e.g., mercury concentration in air = 0.0026 µg/m³, total mercury maximum value in rivers or seas = < 0.0005 µg/l; methylmercury in rivers or seas could not be detected [methylmercury levels in sediments of rivers or seas = < 0.01 µg/g]).

The samples in this study were collected from the customers of the local barbershops of Harbin, Medan, and from each district of Tokushima Prefecture. We chose barbershops randomly from telephone books. The geographical locations included in this study are shown in Figure 1.

Laboratory analysis. Before the analysis, investigators washed hair samples with acetone, rinsed them with distilled water, washed them again with acetone, and then air-dried the samples at room temperature. Researchers cut the dried samples into pieces and weighed them, after which they analyzed them. We used gold-amalgamation cold-vapor atomic absorption spectrometry to measure total mercury levels. We used the ECD-gas chromatography method of Akagi to measure methylmercury levels. To check the accuracy of these two analytical methods used in our laboratory, we, before beginning hair-sample analysis, measured NIES CRM No. 5 human hair to determine total mercury and measured NIES CRM No. 13 human hair to determine methylmercury. The NIES CRM No. 5 and No. 13 human hairs, which were prepared by the National Institute for Environmental Studies, Environmental Agency of Japan, were the human hair certified reference materials for total mercury, methylmercury, and trace-elements analysis. In the NIES CRM No. 5

Fig. 1. Geographical locations of Harbin, Medan, and Tokushima.
human hair, the certified value given for total mercury analysis was $4.4 \pm 0.4 \mu g/g$ dry weight, and the certified value of NIES CRM No. 13 human hair for methylmercury analysis was $3.8 \pm 0.4 \mu g/g$ dry weight. The results we measured in our laboratory for these two NIES hairs are shown in Tables 1 and 2. The analytical results for these two NIES hairs agree well with the certified values provided in the literature. During analysis of all samples, we conducted repeated analysis for the same sample. The analytical results for both total mercury and methylmercury analyses showed good reproducibility.

Prior to analyses, we also discussed the effect of washing hair on hair mercury levels. Although we published a paper about the effect of washing on removal and/or elution of some hair metals in a previous study, we did not, at that time, discuss the effect of washing on hair mercury levels. However, we found no differences between hair total mercury or methylmercury levels either before or after we washed the samples.

**Statistical analysis.** Given that both frequency histograms of total mercury and methylmercury evidenced a nearly logarithmic normal distribution, we converted the data to logarithmic values when we performed the t test of differences of mean values. We used Student's unpaired t test to test the differences between mean values of mercury in hair samples obtained in the different areas. To assess correlation between two groups of variables, we calculated Pearson's correlation coefficients. We set the level of statistical significance at $p < .05$.

**Results**

Total mercury and methylmercury levels in hair samples obtained from the different study areas are shown in Tables 3 and 4. Mercury levels in hair samples differed markedly among the different areas. Mean mercury levels in hair were lowest in China, where subjects had a mean total mercury concentration of 1.694 $\mu g/g$ and a mean methylmercury concentration of 0.416 $\mu g/g$. Subjects in Indonesia had the second highest mean total mercury and methylmercury levels (3.133 $\mu g/g$ and 0.779 $\mu g/g$, respectively), which were approximately 1.8 and 1.9 times the values in Chinese subjects ($p < .01$). Subjects in Japan had the highest mean total mercury and methylmercury levels (4.624 $\mu g/g$ and 3.570 $\mu g/g$, respectively), which were approximately 2.7 and 8.6 times the respective levels in Chinese subjects and 1.5 and 4.6 times the respective levels in Indonesian subjects ($p < .01$).

Mercury levels in hair samples also varied between the different districts of Tokushima Prefecture, Japan. The residents of the mountainous district had the lowest total mercury and methylmercury levels (2.758 $\mu g/g$ and 2.137 $\mu g/g$, respectively). The residents of the middle district had a mean total mercury concentration of 4.069 $\mu g/g$ and a mean methylmercury concentration of 3.150 $\mu g/g$ (i.e., approximately 1.5 times the total mercury and methylmercury mean concentrations determined in the mountainous district [$p < .01$]). The residents of the coastal district had the highest total mercury and methylmercury levels (i.e., 6.245 $\mu g/g$ and 4.917 $\mu g/g$, respectively, on average), which were approximately 2.3 and 1.5 times higher than we found in residents of the mountainous and middle districts, respectively ($p < .01$).

The hair of subjects in the coastal district in Tokushima Prefecture contained approximately 3.7 and 12 times the mean total mercury and methylmercury levels, respectively, of hair provided by subjects in China ($p < .01$). Of the three countries surveyed, we determined that Chinese subjects had the lowest hair mercury levels.

We used histograms to analyze distribution of parameters in each study. The distributions of mercury levels in hair samples, by country, are shown in Figure 2. The distribution of hair mercury levels among the three districts of Tokushima Prefecture, Japan, is shown in Figure

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**Table 1.—Results of NIES CRM No. 5 Human Hair Analysis**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Values of measurements</th>
<th>Recovery ratio (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>4.33</td>
<td>98.41</td>
</tr>
<tr>
<td>2</td>
<td>4.42</td>
<td>100.45</td>
</tr>
<tr>
<td>3</td>
<td>4.36</td>
<td>99.55</td>
</tr>
<tr>
<td>4</td>
<td>4.36</td>
<td>99.09</td>
</tr>
<tr>
<td>5</td>
<td>4.46</td>
<td>101.36</td>
</tr>
<tr>
<td>6</td>
<td>4.43</td>
<td>98.41</td>
</tr>
<tr>
<td>7</td>
<td>4.30</td>
<td>97.73</td>
</tr>
<tr>
<td>$\bar{x} \pm SD$</td>
<td>4.37 ± 0.06</td>
<td>99.29 ± 1.27</td>
</tr>
<tr>
<td>CV</td>
<td>1.37#</td>
<td>1.28</td>
</tr>
</tbody>
</table>

Notes: $\bar{x} =$ mean, $SD =$ standard deviation, and CV = coefficient of variation.
*Certified value: $4.4 \pm 0.4 \mu g/g$ dry weight.
†$\mu g/g$ dry weight.
‡Percentage (%).

**Table 2.—Results of NIES CRM No. 13 Human Hair Analysis**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Values of measurements</th>
<th>Recovery ratio (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>3.76</td>
<td>98.95</td>
</tr>
<tr>
<td>2</td>
<td>3.83</td>
<td>100.79</td>
</tr>
<tr>
<td>3</td>
<td>3.69</td>
<td>97.31</td>
</tr>
<tr>
<td>4</td>
<td>3.78</td>
<td>99.47</td>
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<tr>
<td>5</td>
<td>3.81</td>
<td>100.26</td>
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<tr>
<td>6</td>
<td>3.72</td>
<td>97.89</td>
</tr>
<tr>
<td>7</td>
<td>3.80</td>
<td>100.00</td>
</tr>
<tr>
<td>$\bar{x} \pm SD$</td>
<td>3.77 ± 0.05</td>
<td>99.21 ± 1.32</td>
</tr>
<tr>
<td>CV</td>
<td>1.33#</td>
<td>1.33</td>
</tr>
</tbody>
</table>

Notes: $\bar{x} =$ mean, $SD =$ standard deviation, and CV = coefficient of variation.
*Certified value: $3.8 \pm 0.4 \mu g/g$ dry weight.
†$\mu g/g$ dry weight.
‡Percentage (%).
Table 3.—Total Mercury Concentrations (μg/g) in Hair of Subjects in Different Countries and Districts of Tokushima

<table>
<thead>
<tr>
<th>Population area</th>
<th>̄x</th>
<th>SD</th>
<th>Min.</th>
<th>Max.</th>
<th>n</th>
</tr>
</thead>
<tbody>
<tr>
<td>Harbin (China)</td>
<td>1.694</td>
<td></td>
<td>4.979</td>
<td>36.356</td>
<td>64</td>
</tr>
<tr>
<td>Medan (Indonesia)</td>
<td>3.133</td>
<td></td>
<td>4.697</td>
<td>19.888</td>
<td>55</td>
</tr>
<tr>
<td>Tokushima (Japan)</td>
<td>4.624</td>
<td></td>
<td>2.753</td>
<td>24.644</td>
<td>243</td>
</tr>
<tr>
<td>Mountainous district</td>
<td>2.758</td>
<td></td>
<td>1</td>
<td>5.642</td>
<td>39</td>
</tr>
<tr>
<td>Middle district</td>
<td>4.069</td>
<td></td>
<td>3.333</td>
<td>9.994</td>
<td>126</td>
</tr>
<tr>
<td>Coastal district</td>
<td>6.245</td>
<td></td>
<td>3.717</td>
<td>24.644</td>
<td>78</td>
</tr>
</tbody>
</table>

*p < .01.

Table 4.—Methylmercury Concentrations (μg/g) in Hair of Subjects in Different Countries and Districts of Tokushima

<table>
<thead>
<tr>
<th>Population area</th>
<th>̄x</th>
<th>SD</th>
<th>Min.</th>
<th>Max.</th>
<th>n</th>
</tr>
</thead>
<tbody>
<tr>
<td>Harbin (China)</td>
<td>0.416</td>
<td></td>
<td>0.111</td>
<td>1.271</td>
<td>64</td>
</tr>
<tr>
<td>Medan (Indonesia)</td>
<td>0.779</td>
<td></td>
<td>0.143</td>
<td>2.762</td>
<td>55</td>
</tr>
<tr>
<td>Tokushima (Japan)</td>
<td>3.57</td>
<td></td>
<td>0.197</td>
<td>17.5</td>
<td>243</td>
</tr>
<tr>
<td>Mountainous district</td>
<td>2.137</td>
<td></td>
<td>0.992</td>
<td>4.736</td>
<td>39</td>
</tr>
<tr>
<td>Middle district</td>
<td>3.15</td>
<td></td>
<td>0.197</td>
<td>7.04</td>
<td>126</td>
</tr>
<tr>
<td>Coastal district</td>
<td>4.917</td>
<td></td>
<td>1.279</td>
<td>17.5</td>
<td>78</td>
</tr>
</tbody>
</table>

*p < .01.

Fig. 2. Distribution of mercury levels in hair samples for each country.
3. As the mean mercury level decreased, the parameter tended to be distributed at low levels on the histogram; as the mean mercury level increased, the parameter tended to be distributed at higher levels on the histogram. The percentage of subjects who had a total mercury level of 1 µg/g or less was 79.7% in China, 30.9% in Indonesia, and 0.8% in Japan. Most subjects in China had a total mercury level below 1 µg/g, whereas most subjects in Japan had a total mercury level that exceeded 1 µg/g. The percentage of subjects who had total mercury levels that exceeded 8 µg/g was 3.13% in China, 12.7% in Indonesia, and 7% in Japan. When we conducted a similar analysis for methylmercury levels, the percentage of subjects who had methylmercury lev-
levels below 1 μg/g was 96.9% in China, 76.4% in Indonesia, and 1.7% in Japan. Most subjects in China and Indonesia had a methylmercury level over 1 μg/g. The percentage of subjects who had methylmercury levels over 8 μg/g was 0% in China and Indonesia and 2.9% in Japan. The maximum methylmercury level was 1.271 μg/g in China and 2.762 μg/g in Indonesia. Of all the Japanese subjects, 59.7% had methylmercury levels higher than the maximum level we found in Indonesia.

Among the three survey districts of Tokushima Prefecture, total mercury and methylmercury levels in residents of the mountainous district were distributed in the low end of the range, levels in residents of the middle district were distributed in the middle of the range, and levels in residents of the coastal district were distributed in the highest portion of the range (Fig. 3).

The correlation between hair total mercury and methylmercury levels in individual subjects, by country, is shown in Figure 4. In Tokushima, Japan, an individual’s total mercury level correlated very closely with his or her methylmercury level (r = .937). The correlation between those two parameters was lower in China and Indonesia (i.e., methylmercury levels in Chinese and Indonesian subjects were not high—even if the total mercury levels were high). All three Japanese districts were correlated closely with respect to hair total mercury and methylmercury levels (Fig. 5). Close correlation between these two parameters was also seen in the coastal district, where the total mercury level was often high.

Discussion

Some investigators reported that hair mercury levels in residents of some areas contaminated with environmental mercury (i.e., induced by human activity) were higher than levels in residents of areas absent mercury contamination. In 1995, Akagi reported that in the Tapajos area of the Amazon River Basin (Brazil)—an area of mercury pollution that has resulted from gold-mining activities—residents of fishing villages were more exposed to methylmercury than people who lived far from this area. Investigators tested several hair samples, and they found methylmercury levels that exceeded 50 ppm. On the other hand, hair samples from goldminers and workers in goldshops contained relatively high levels of mercury, which were mainly in inorganic form. Leino and Lodén performed a similar study in the Tucuruí area in Brazil, and they found that, in the area contaminated by mercury, hair mercury levels of fishermen and their families ranged from 0.9 to 240 ppm (mean = 65 ppm). Hair mercury levels among the fishermen and their families in Tucuruí area were sufficiently high to cause health effects.

Investigators have also studied hair mercury levels of subjects from districts that are characterized by a lack of contamination induced by human activity. Suzuki analyzed mercury levels in hair samples obtained from three villages of Papua, New Guinea: (1) Wonie (approximately 25 km from the coast); (2) Ume (on the bank of the Binaturi River, approximately 6 km from the coast); and (3) Dorogori (on the coast). Mean hair mercury levels were highest in Dorogori (males = 4.1 μg/g, females = 4.4 μg/g) and were lowest in Wonie (males = 1.5 μg/g, females = 1.0 μg/g); levels in Ume were 3.8 μg/g for males and 3.4 μg/g for females. In the study by Bruhn et al., they determined total mercury content in scalp hair of (a) 59 pregnant and nursing women (i.e., normal to high consumption of fish and seafood) who resided in fishing villages throughout the coastal zone of the Eighth Region of Chile; and (b) 7 pregnant and nursing women (i.e., negligible or no fish/seafood consumption) who resided in Pinto, an inland town in the same region. They reported a mean total mercury content in hair of the study group of 2.06 μg/g, which was significantly higher than in the control group (0.43 μg/g).

Our results also indicated that mercury levels in hair samples differed markedly among the three countries surveyed, and mean mercury levels in hair were lowest in Harbin, China (approximately 600 km from the coast). Subjects in Medan, Indonesia (approximately 30 km from the coast) had the second highest mean mer-
cury levels. Subjects in Tokushima Prefecture, Japan (contiguous to the Pacific Ocean), had the highest mean mercury levels. Mercury levels were lowest in Harbin, China; higher in Medan, Indonesia, and were the highest in Tokushima, Japan. Among the three districts of Tokushima, Japan, residents in the mountainous district—which was farther from the coast than the middle and coastal districts—were engaged primarily in agriculture or forestry activities, and they had the lowest mercury levels. Residents in the middle district, which was located between the mountainous and coastal districts, had the second highest mean mercury levels. Residents in the coastal district, which faced the Pacific Ocean, earned their living primarily via a mixture of agricultural and fishing activities, and these individuals had the highest mean mercury levels. In the three districts of Tokushima, mercury levels were highest in the coastal district, followed by individuals in the middle district: levels were lowest in the mountainous district. In Tokushima, Japan, an individual's total mercury level correlated very closely with his or her methylmercury level; however, the correlation between these two parameters was lower in Harbin, China, and in Medan, Indonesia. Methylmercury levels in Chinese and Indonesian subjects were not high—even if their total mercury levels were high.

Regional differences in hair mercury levels of Japanese people appeared to reflect differences in the amounts of fish and shellfish ingested. In a survey done by the Japan General Affairs Department in 1996, investigators reported that fish and shellfish consumption in Japan (106.9 g/d · person) was the third highest in the 23 countries surveyed. In a 1994 survey from Tokushima Prefecture, researchers reported consumption of 110.4 g/d · person in this area. In the different districts of Tokushima, the levels of consumption in the mountainous, middle, and coastal districts were 100.8 g/d · person, 105.6 g/d · person, and 139 g/d · person, respectively. On the basis of these results, we estimated that methylmercury in fish and shellfish was a major source of mercury detected in the hair of Japanese people. In contrast, the low levels of mercury in hair—especially the low methylmercury levels for residents in Harbin, China—are likely explained by the fact that people in this city, located far from the coast, ingest very little fish.

Mercury and its compounds occur naturally in the environment, but their use in industry and their release into the atmosphere via burning of fossil fuels and processing of ores can increase ambient environmental levels. Occupational exposure to mercury results mainly from exposure to elemental mercury vapor. Some examples of people who are exposed occupationally include chemical industrialists, pesticide preparers, dentists, dental amalgam filling makers, workers in the chloralkali industry, metal-refining and fluorescent-tube factories, and thermometer manufacturers. In most of these workplaces, inorganic mercury vapor is released; however, in some countries there are recommended maximum levels for working areas, but individuals are nonetheless exposed because they are ignorant of the inherent dangers or because the companies are unable to reduce levels in old plants.

Medan, the fourth largest city in Indonesia, is an important industrial center that includes an oil field. The major industries in Medan are chemical factories, which produce urea-based fertilizers and petrochemicals. Harbin, which is located in northeast China, is also an important industrial city. High total mercury levels (note: methylmercury levels were not high) from the hair of some subjects in Harbin and Medan have resulted from increased exposure to inorganic mercury that results from an occupational or environmental source; however, we did not know the occupations of these subjects.

Our results highlight the necessity of considering regional characteristics (including regional differences in diet) when hair samples are used in the biological monitoring of mercury.

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Requests for reprints should be sent to Dr. Qiuyang Feng, Department of Hygiene, School of Medicine, Tokushima University, Kuramoto-cho, Tokushima 770, Japan.

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References


