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Mercury concentration in hair samples from Chinese people in coastal cities

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Abstract

This investigation was made to estimate current normal concentrations of total mercury in the hair of Chinese coastal people. Hair samples were collected from 659 healthy inhabitants in the areas along the coast and the rivers (such as Shanghai, Ningbo, Dalian, Xiamen, and Zhoushan) of China from Feb. 2005 to June 2006. Total mercury concentrations in the samples were analyzed by the cold vapor atomic adsorption spectrometry method (CVAAS). The results showed the geometric mean concentration of total mercury in the hair of the total population (N = 659) was 0.83 μg/g, and the geometric mean of the concentrations of total mercury in the hair of the male (0.94 μg/g, 338 inhabitants with an average age of 35.2, age 2–93) was higher than that of the female (0.72 μg/g, 321 inhabitants with an average age of 39.1, age 1–90). In both the male and female, the mean concentrations of total mercury in hair increased with age, and then gradually decreased. There was a significant correlation between the total hair mercury and the place of residence ($p <$ 0.01), and total hair mercury was the highest in volunteers who lived in Zhoushan. Over half of all sample concentrations were below the USEPA-recommended 1 μg/g. In Zhoushan (males, 2.44 μg/g; females, 1.94 μg/g) and Ningbo (males, 1.06 μg/g; females, 1.02 μg/g), it exceeded the recommended level. Very little females (0.3%) of reproductive age showed hair mercury levels close to 10 μg/g. It was thus concluded that fish consumption was a major current route for mercury exposure in China.

Key words: hair mercury; Chinese people; coastal cities

Introduction

Mercury is a hazardous metal that in the last decades had caused serious episodes of environmental contamination and human intoxication in several locations around the world, including the Minamata outbreaks (Harada, 1995). The potential risk of health hazards of humans by mercury exposure has been assessed by estimating the metal contents in breast milk, blood, hair, nail, adipose tissues, and various organs (Apostoli *et al*., 2002; Clarkson 2002; Minoia *et al*., 2007). Among them, however, total hair mercury should be the best indicator. Compared to other specimens, hair has several advantages in that it may be obtained by non-invasive sampling and contains higher concentrations of elements. In addition, tracing the dynamic changes in the hair growing direction provides an insight into average exposure on a time-scale ranging from weeks to months. Moreover, excellent analytical methods have been established including cold flameless atomic absorption spectrophotometry using gold amalgam, which gives good precision and reproducibility. Therefore, scalp hair has been used as a biomarker in many mercury exposure studies (Yasutake *et al*., 2003; Pinheiro *et al*., 2005; Sakamoto *et al*., 2008).

China is one of the countries with the greatest mercury production in the world. The major sources of mercury in the environment in China have been Hg production and coal burning. The biological health effects of mercury pollution in mercury mines have been reported (Cheng *et al*., 2005a, 2005b, 2005c, 2006a; Sakamoto *et al*., 2007a). High levels of mercury in the food and air of China cities have been reported in several studies (Horvat *et al.*, 2003; Cheng *et al*., 2005c, 2006b). However, studies on mercury concentrations in the hair of Chinese people have been quite limited. The present study was made to estimate the current normal level of mercury in the hair of the Chinese living in coastal cities. We analyzed the hair mercury levels at varying ages among the general population in five China coastal districts. Considering the geological characteristics, five sampling areas were selected in this study.

1 Materials and methods

1.1 Hair sampling

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Six hundred and fifty-nine hair samples (healthy volunteers, 60–100 hairs, cut as close to the scalp as possible) were collected in the cities of Dalian Lvshun, Shanghai, Ningbo, Zhoushan, and Xiamen from Feb. 2005 to June 2006, while regarding the age and sex. Collected hair samples were kept in a clean polyethylene bag until chemical analysis. Mean age and its range were 35.2 and 2–93 for males, and 39.1 and 1–90 for females, respectively.

1.2 Mercury analysis

For mercury analysis, the hair samples were washed with detergent, and rinsed two times with acetone to dry. The dried hair was cut into small pieces shorter than 2 mm with scissors. Mercury contents in the samples were analyzed using the cold vapor atomic adsorption spectrometry method (CVAAS, HG-201, Japan) according to the method of Akagi *et al*. (2000), which involves sample digestion with HNO_3, $HClO_4$, and H_2SO_4 (1+1+5), followed by reduction of divalent Hg to Hg^0 vapor with $SnCl_2$ with the detection limit of 0.01 ng/g (Sakamoto *et al*., 2008).

1.3 Statistical analysis

Statistical evaluation of the data was performed using a SPSS 11.0 statistical package.

2 Results

2.1 Frequency distribution, range and geometric mean

Frequency distributions of the mercury level of hair samples collected in the five districts are summarized in Fig.1. This figure showed the mercury concentrations were apparently distributed in a lognormal manner, so the geometric mean was used as representative of hair mercury levels instead of the arithmetic mean in this study. The frequency distributions of the mercury concentrations were the same in males and females. The results were similar with those reported by Yasutake *et al*. (2003). The range and geometric mean of the mercury level of all hair samples are shown in Table 1, in which the hair mercury concentration in the total population (N = 659) was 0.83 μg/g. The hair mercury concentration level was higher in the total male population (N = 338) with a geometric mean of 0.94 μg/g than in the total female population (N = 321) with geometric mean of 0.72 μg/g.

2.2 Relation between age and hair mercury

The relation between age and concentration of total hair mercury in males (338 healthy volunteers ranging 2–93 years of age, average age 35.2) is shown in Fig.2. The total hair mercury was almost constant from inhabitants in their teens to twenties, increased from the lowest mean values in the twenties to forties to the highest mean value, and then gradually decreased with age. The total hair mercury was almost constant after the seventies.

Figure 3 shows the relation between age and total hair mercury in females (321 healthy volunteers 1–90 years of age, average age 39.1), and the mean total hair mercury in each age group was shown by a solid line. The total hair mercury decreased from the lowest mean values in the teens to twenties, and then gradually increased to the highest mean values in the thirties. The total hair mercury was almost constant after the forties.

2.3 Sexual differences and correlation between total hair mercury and place of residence

The sexual differences were not so apparent as shown in Fig.4, especially in Ningbo City. The age-dependent

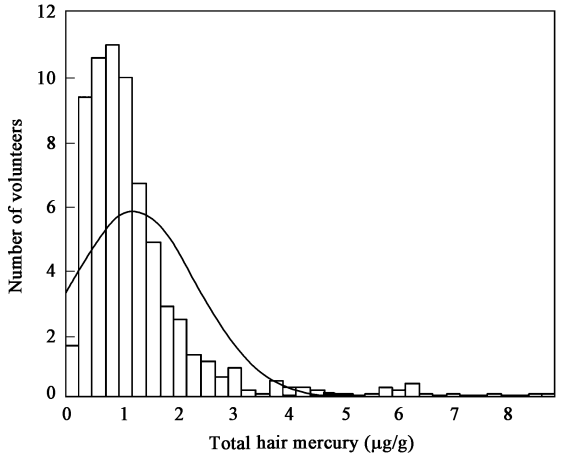

Fig. 1 Frequency distribution of total hair mercury in 659 volunteers.

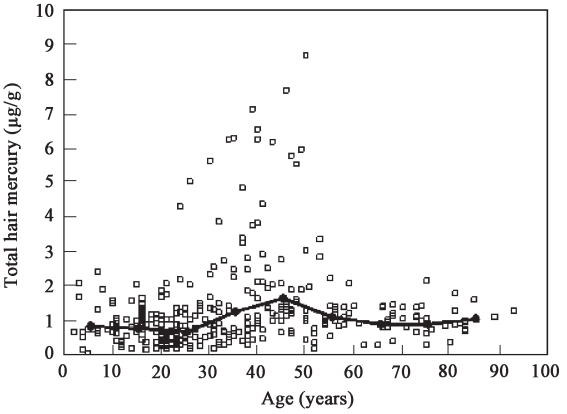

Fig. 2 Relation between age and total hair mercury in males. Solid line represents the means of total hair mercury in respective age groups.

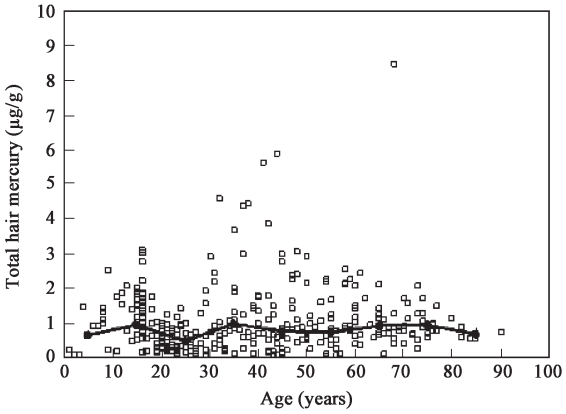

Fig. 3 Relation between age and total hair mercury in females. Solid line represents the means of total hair mercury in respective age groups.

Table 1 Geometric mean and range of the hair mercury content in five geological populations

Sampling station	Sex	Number of volunteers	Age (years)	Geometric value (μg/g)	Range (μg/g)
Xiamen City	Male	14	2–76	0.78	0.28–3.85
	Female	28	21–54	0.93	0.21–2.27
Zhoushan City					
	Male	50	7–53	2.44	0.43–8.70
	Female	47	15–73	1.94	0.75–5.88
Ningbo City	Male	147	3–93	1.06	0.32–5.79
	Female	115	1–90	1.02	0.22–2.98
Shanghai City	Male	88	7–62	0.60	0.16–4.31
	Female	75	16–84	0.41	0.06–4.59
Dalian Lvshun City	Male	39	4–80	0.52	0.03–2.72
	Female	56	2–76	0.29	0.04–8.48

alterations in the five districts were quite different from each other. Contrary to the other cities, in Xiamen City the female mercury concentration levels were higher than the male levels. There was a very significant correlation between the total hair mercury and the place of residence ($p < 0.01$). The alteration of the mercury levels with age in Zhoushan City was more apparent than the others. The geometric mean was significantly higher in Zhoushan City, 2.44 μg/g in males and 1.94 μg/g in females, than in the others ($p < 0.01$). The age-dependent distribution profile obtained in Zhoushan City was slightly different from those of the other four sampling sites, reflecting a relatively higher exposure level to methylmercury in most age classes.

2.4 Cumulative frequency of individual hair mercury content

Table 2 indicates the cumulative frequency of the individual hair mercury content among all samples. Over half of the population possessed hair mercury contents below the USEPA-recommended 1 μg/g. It was also found that the hair levels of small portions of the subject population, 0.3% females, were close to 10 μg/g, which is a critical hair mercury level with possible adverse effects on the developing fetus in uterus (Sakamoto *et al*., 2002; Yasutake *et al*., 2003).

3 Discussion

For children, mercury can affect the development of the nervous system. While fish consumption is often encouraged to increase gestation in pregnancy and bolster developing children's neurodevelopment, high mercury exposure in mothers may lead to very preterm delivery and counterproductive effects on children's neurodevel-

Table 2 Cumulative frequency of individual hair mercury content (%)

Sex	Age (years)	Total	Mercury concentration (μg/g)				
			< 1	< 2	< 3	< 5	< 10
Female	All	321	62.9	87.9	96.3	98.8	100
	15–49	200	64.5	88.0	94.5	98.5	100
Male	All	338	52.1	85.8	92.9	95.9	100
Total	All	659	57.45	86.8	94.5	97.3	100

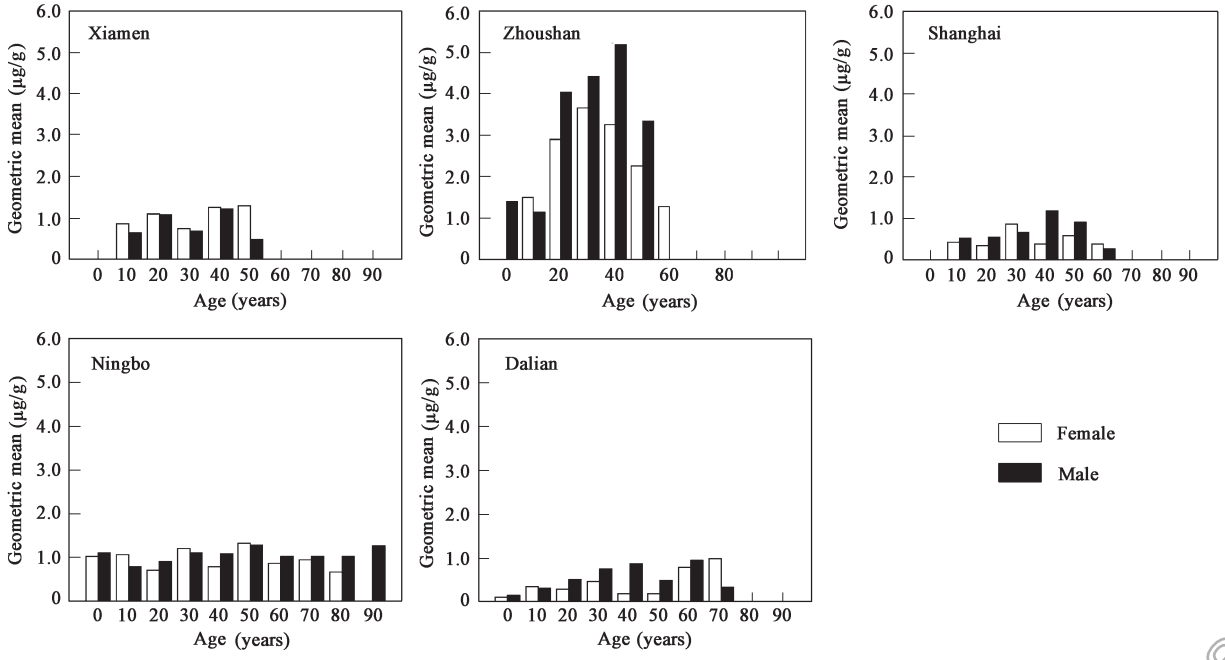

Fig. 4 Age-dependent distribution of geometric mean of hair mercury content in Xiamen, Zhoushan, Ningbo, Shanghai, and Dalian cities.

opment. For adult men, there is emerging evidence that elevated mercury levels (hair mercury greater than 2 μg/g) may cause an increased risk of cardiovascular disease. In this study, T-Hg levels were determined in scalp hair of residents in five coastal cities of China. Different T-Hg concentrations in the five cities were mainly dependent on age, gender, and dietary customs. These hair Hg values provide an estimate of exposure over an approximate 2-month period, as recent exposure is not yet incorporated into the hair growth outside of the scalp. The mean total hair Hg levels (0.83 μg/g) of these cities were generally lower than the levels reported in other studies of China and international populations; for example, the mean hair Hg of Chinese mercury mine workers was 20.0 μg/g (Sakamoto *et al*., 2007a), and the geometric mean of Japanese residents in five districts was 2.55 μg/g for males and 1.43 μg/g for females (Yasutake *et al*., 2003). Previously we found that hair mercury levels in Ningbo City in 2000 were much higher than the present levels (mean = 1.72 μg/g, n = 48; 1.06 μg/g for males and 1.02 μg/g for females). This could be attributed to several factors. One may be that mercury levels in China are actually lower than they were 6 years ago. On the other hand, this survey might simply have missed many of the people who presently eat more fish containing much mercury.

Many studies showed that gender was unlikely to play a role in determining mercury accumulation in hair (Kosatsky *et al*., 2000; Mortada *et al*., 2002; Olivero *et al*., 2002). Nevertheless, in this study, female hair was found to have a lower mean (0.72 μg/g) mercury level than male hair (0.94 μg/g). Moreover, these differences were significant. Similar findings were reported elsewhere (Yasutake *et al*., 2003; Díez *et al*., 2008). The relationships between Hg concentrations and age were tested (Figs.2 and 3). No clear, statistically distinguishable trends were observable, although the mean concentration of total Hg increases with age up to forties for males and thirties for female, then decreases thereafter. Buzina *et al*. (1995) also found that the Hg content of hair did not increase significantly after the age of 35 years. Although, Bou-Olayan and Al-Yakoob (1994) found a weak correlation between Hg contents and the age of the donor (r = 0.44), these findings may have been influenced by not removing grey hairs from the samples before the analysis, as grey hairs have no Hg content (Al-Majed and Preston , 2000).

The hair mercury level in Zhoushan City was relatively higher (mean 2.44 μg/g for males and 1.94 μg/g for females) than the levels estimated from hair mercury concentrations observed in the other four districts. The positive result obtained suggested that fish consumption was the main exposure route for mercury. The age-related fluctuation in total hair mercury found in Zhoushan City might also have partially resulted from the difference in food consumption. This area faces the Pacific Ocean, there are many fisherman here, and the price of fish is much lower than in other cities. Moreover, the dietary customs in Zhoushan City are difference from other cities. Fillets are common in every course in Zhoushan City, therefore, high fish consumption would be expected. Most of the fish they consume was purchased and prepared at home or eaten in a restaurant. While fish and shellfish provide a high protein, low-fat alternative to meat and poultry, frequent consumption of large predatory fish can pose a risk of exposure to persistent environmental contaminants such as organochlorines and methylmercury which bioaccumulate in the aquatic food chain.

Based on the data in Minamata, Niigata and Iraq, it was reported earlier that no health effect was observed with hair mercury levels below 50 μg/g for adults. However, since a fetus is most vulnerable to MeHg toxicity (Sakamoto *et al*., 2002, 2007b), the report also suggested that the mercury levels of pregnant women should be kept less than 10 μg/g (WHO, 1990). The United States Environmental Protection Agency postulated 0.1 μg/(kg body weight·d) as a reference dose (RFD) of methylmercury (USEPA, 1997), an ingestion dose limit that is considered to cause no adverse health effect to a human, including sensitive subpopulations. The USEPA's RFD level corresponds to a hair mercury concentration of 1 μg/g (USEPA, 1997). In this study, 57.45% of all samples were below the USEPA-recommended 1 μg/g. However, this did not mean that all the people were exposed to the safe level. In Zhoushan (males, 2.44 μg/g; females, 1.94 μg/g) and Ningbo (males, 1.06 μg/g; females, 1.02 μg/g), the recommended level was exceeded, and very few (0.3%) females at the reproductive age showed hair mercury levels close to 10 μg/g. Because no questions about dietary habit were including in our questionnaire, the analysis of relationship between hair mercury and fish consumption could not be conducted. Based on the past research, dietary fish/shellfish was a major mean of people expose to the contaminant (Nakagawa, 1995; Batista *et al*., 1996). In most countries, fish is the most important mercury source for the general population (Horvat and Gibičar, 2006). In comparison with other countries, Chinese fish/shellfish consumption had increased greatly from 1982 to 2002, according to a large portion of world fish consumption. Thus, we could still considered that the different amounts and species of fish consumed led to varying hair mercury levels in the five coastal areas. With fish consumption and greater mercury emissions in the environment, more and more residents especially in Chinese coastal areas run the risk of exposure to methylmercury.

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