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Changes in hair arsenic concentration in a population exposed to heavy pollution: Follow-up investigation in Chenzhou City, Hunan Province, Southern China

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Abstract

Follow-up investigation on hair arsenic concentration was conducted in an arsenic heavily polluted area of southern China in 2002 and 2006. The results showed that the geometric mean of hair arsenic concentration decreased from 2.95 mg/kg in 2002 to 1.78 mg/kg in 2006, when the percentage of the population with levels over 1 mg/kg only decreased from 93.4% in 2002 to 80.5% in 2006. Over this four-year period, the population with high arsenic concentrations decreased significantly while there was no obvious change in hair arsenic concentration for people who had relatively low concentrations. In terms of age distribution, young and old people had higher hair arsenic concentrations than the middle-aged. All of these results showed that it is difficult to reverse the negative impact of arsenic pollution on human health. Arsenic pollution has a long-term continuous influence on the health of local residents.

Key words: arsenic; human health; soil contamination; hair arsenic

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Introduction

Arsenic is one of the worst cancer-causing chemicals distributed widely in the environment from both natural and anthropogenic sources (Moon et al., 2004; Wang and Duan, 2009). In soil it can a pose risk to human health either by ingestion via the food chain or through secondary pollution of air and water due to dust and leaching loss (Zeng et al., 2008). It is estimated that 30%–88% of the As absorbed by humans is from As-polluted soil (Diazbarriga et al., 1993). The levels of accumulated As in humans can be evaluated by measuring As concentration in the hair (Agusa et al., 2006; Bencko, 1995; Kurttio et al., 1998; Saad and Hassanien, 2001), as this is highly correlated with As absorption (Kazi et al., 2009). Hair concentrations of As reflect the amount of As absorbed into the human body within the several months preceding hair sampling (Akagi et al., 1995; Boischio and Cernichiari, 1998; Gault et al., 2008). Since 1970, hair has been utilized in numerous human studies to reveal chronic As exposure, providing useful information on chronic As poisoning (Bencko, 1995). Hair analysis provides convenient sampling, easy sample conservation and transportation, and is non-invasive to human health (Pragst and Balikova, 2006; Qin, 2004).

There are some research works on the hair As concentrations of people living in polluted areas. However, follow-up studies on the changes of hair As concentration in As polluted areas is limited. In this study, we conducted a follow-up investigation on hair As concentrations to analyze changes of the hair As concentration within a four-year interval (2002 and 2006), and evaluate the continuous impacts of As contaminated soil on human health.

1 Materials and methods

1.1 Study area

Continuous discharge of As from Dengjiatang Arsenic Factory in Chenzhou City, Hunan Province, China polluted the surrounding environment and made hundreds of the residents to be hospitalized for medical treatment in 2000 (Liao et al., 2003). The paddy and vegetable fields around the factory were heavily polluted with As, with concentrations in the grains and vegetables (e.g., cabbage, radish and spinach) significantly exceeding the National Standards of Food Safety (Cai et al., 2004). Consequently, the villagers influenced by As pollution began to consume clean tap water to replace polluted well water and the polluted farmlands were abandoned. The soil As concentration in the study area ranged from 30.9 to 255 mg/kg with an average value of 97 mg/kg. The As concentrations in the farm fields were multiples of the limit set in Standard for Environmental Quality of Soil (GB 15618-1995).

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1.2 Study cohort

People investigated are residents living in the As polluted areas for a long time, and dieted on polluted crops before the As pollution. They stopped to eat local crops and turned to diet foods from markets after the As pollution and use clean tap water which satisfy the As standard. In 2002, 76 residents (age range 0–76 year) were recruited from the study area controlling for gender. For comparison, four years later in 2006, 82 residents (age range 0–76 year) were recruited from the same area, in which 26 of residents were investigated in 2002.

1.3 Sampling and analysis

Approximately 1.0 g of newly grown hair was cut from the neck nape of each resident using stainless steel scissors. Only hair segments of 3 cm length closest to the scalp were collected. The hair samples were immediately stored in individual sealable polyethylene sample bags to avoid contamination.

All samples were washed with shampoo to remove any surface contamination, rinsed in deionized water, and then air dried in an oven at 50°C for 4 hr. The dried hair was cut into small pieces (< 1 mm) using stainless steel scissors, and the pieces were stored in polyethylene tubes at room temperature until analysis. Samples were weighed immediately prior to digestion.

Hair samples were digested with a mixture of HNO_3 (10 mL) and $HClO_4$ (2 mL) in electrothermal heating blocks until the perchloride was almost driven off. Arsenic concentration was determined using a hydrogen generation-atomic fluorescence spectrometer (HG-AFS) (AFS-2202, Haiguang Instrumental Co., China) (Liao et al., 2005). Shampoo was also evaluated and As was not detected in it.

Reagent blank and human hair certified reference material (GBW 07601) from the National Research Center for Standard Materials (Beijing, China) were used to verify the analytical procedures. This reference material contains a certified total As concentration of (0.28 ± 0.05) mg/kg.

1.4 Statistical analysis

The data were statistically analyzed using SPSS 17.0. The level of significance was set at $p < 0.05$.

2 Results

2.1 Arsenic concentrations in human hair

For quality control, the total As was determined in the hair certified reference material (CRM) at (0.30 ± 0.04) mg/kg (n = 3), which agrees with the certified total concentration of CRM (0.28 ± 0.05) mg/kg. The CRM was not washed prior to the total As analysis.

Kolmogorov-Smirnov (K-S) test of the hair As concentration data showed a log-normal distribution. The geometric mean value of total As concentration was 2.95 mg/kg in 2002 and 1.78 mg/kg in 2006. Independent sample t-tests and LSD multiple-comparison analysis of variance were calculated for hair As concentrations in groupings of different gender, occupation (peasant or non-peasant) and age. The statistical analysis results are summarized in Tables 1, 2 and 3.

Independent sample t-tests with a logarithmic transformation showed that hair As concentrations declined significantly from 2002 to 2006 ($p < 0.001$). The geometric mean, the median and the percentage of hair samples with As concentrations over the standard limit (over-standard percentage) decreased 1.17 mg/kg, 0.86 mg/kg and 12.9% (Tables 1 and 2), respectively. The rates of decline for these parameters were 39.7%, 32.0%, 13.8%, respectively. However, the decrease in the over-standard percentage was only 1/3 of the absolute value.

Comparing As concentrations in 2006 with that in 2002, the male geometric mean and median were lower but the

Table 1 Hair As concentrations of residents grouped by age from Dengjiatang As-polluted area

Age group	Number of samples	Concentration of hair As (mg/kg)				Hair sample containing As 1 mg/kg (%)
		Geometric mean	Median	Minimum	Maximum	
Year 2002						
0–9	4	3.41	3.57	2.53	4.23	100
10–19	17	2.67	2.78	0.52	10.83	88.2
20–29	6	2.03	2.32	0.64	5.74	83.3
30–39	18	3.08	2.40	0.61	10.83	94.4
40–49	16	3.47	3.38	1.53	9.86	100
50–59	13	2.77	2.23	0.48	8.62	92.3
> 60	2	4.51	4.67	3.44	5.90	100
Total	76	2.95	2.69	0.48	10.83	93.4
Year 2006						
0–9	14	3.22[a]	3.94	0.93	7.35	92.9
10–19	14	1.65[bc]	1.56	0.45	4.50	85.7
20–29	17	1.06[c]	1.29	0.27	3.27	58.8
30–39	13	1.77[b]	2.06	0.43	8.25	69.2
40–49	10	1.54[bc]	1.54	0.77	3.27	90.0
50–59	10	2.03[ab]	1.89	0.78	5.84	90.0
> 60	4	2.69[ab]	2.83	1.47	4.71	100
Total	82	1.78	1.83	0.27	8.25	80.5

[a,b,c] mean the result of multiple-comparison analysis for As amount in hairs of different age groups (LSD).

Table 2 Hair As concentrations of residents, grouped by gender, from Dengjiatang As-polluted area

Sex	Number of samples	Concentration of hair As (mg/kg)				Hair sample containing As 1 mg/kg (%)
		Geometric mean	Median	Minimum	Maximum	
Year 2002						
Female	42	3.65*	3.80	0.61	10.83	97.6
Male	34	2.27	2.17	0.48	7.10	88.2
Total	76	2.95	2.69	0.48	10.83	93.4
Year 2006						
Female	43	1.49	1.76	0.27	7.35	72.1
Male	39	2.16**	2.02	0.44	8.25	89.7
Total	82	1.78	1.83	0.27	8.25	80.5

* Hair As concentration is significantly different between females and males in 2002 ($p = 0.002$);
** hair As concentration is significantly different between females and males in 2006 ($p = 0.023$).

Table 3 Hair As concentrations of residents, grouped by occupation, from Dengjiatang As-polluted area

Occupation	Number of samples	Concentration of hair As (mg/kg)				Hair sample containing As 1 mg/kg (%)
		Geometric mean	Median	Minimum	Maximum	
Year 2002						
Non-peasants	50	2.44	2.35	0.48	10.83	90.0
Peasants	26	4.26*	4.38	1.69	10.83	100
Total	76	2.95	2.69	0.48	10.83	93.4
Year 2006						
Non-peasants	46	2.05	2.13	0.44	8.25	82.6
Peasants	36	1.49	1.79	0.27	5.84	77.8
Total	82	1.78	1.83	0.27	8.25	80.5

* Hair As concentration is significantly different between farming and non-farming residents ($p = 0.001$).

over-standard percentage increased slightly. However, the corresponding data for the female grouping all showed decreases. In the different age groups all data presented decreases, except the 0–9 year group with the rising in median and the over 60 year group remaining stable in terms of over-standard percentage. The relevant statistics for peasant and non-peasant groupings also decreased.

Hair As concentrations between the various age groups were found to be significantly different in 2006. The 20–29 year group had the lowest concentrations, and the 0–9 year group was higher than others (Table 1). Differences in hair As concentration between the age groups were not significant in 2002. However, from a geometric point of view, the 20–29 year group has the lowest levels and the 40–49 year and over 60 year groups have higher levels than others. Hair As concentrations in non-peasants were significantly lower than peasants in 2002, while there was no significant difference in 2006.

2.2 Changes in hair As concentration in population

There are 26 people being investigated in both 2002 and 2006. In this section, we analyzed the decreasing rate of the hair concentration of every person in this group. A paired *t*-test for hair As concentration in residents recruited in 2002 and 2006 showed a significant difference between the two groups ($p = 0.002$). The rate of decline ($(As_{2002}-As_{2006})/As_{2002}$) for each individual's hair As concentration in these 26 residents was summarized and grouped based on population attributes (Table 4). By K-S test, the rate of decline of hair As concentration was determined to follow a normal distribution.

Over all the age groups the mean decline rate observed was 52%, with a highest maximum of 95% and a lowest minimum of 10%. No significant difference was found between the different genders, ages or occupations. A comparison of mean decline rates between age groups showed that the rate for the middle-aged group (20–49 year) was higher than that of the young group (0–19 year) or the old group (over 50 year) (Table 4). Correspondingly, the individuals with high decline rate (whose decline rate > 50%) were all within the 10–59 year group, meanwhile the highest decline rate was 40% in the 0–9 year group or 37% in the over 60 year group (Table 4).

3 Discussion

3.1 Threshold value for As concentration in human hair

An official standard limit for As in human hair has not been established. Of 1250 human hair samples in the United Kingdom, 80% contained less than 1 mg/kg of As, suggesting that hair As concentrations under 1 mg/kg are not from As poisoning (Hamilton, 1964). Arsenic concentrations in human hair from a selection of non-contaminated and contaminated areas in the world indicate that concentrations in non-contaminated areas, except for Brazil, are typically lower than 1.0 mg/kg (Table 5). A value greater than 1 mg/kg is considered to indicate chronic exposure and toxicity (Hindmarsh, 2000, 2002;

Table 4 Rate of decline in hair As concentration of residents in Dengjiatang As-polluted area from 2002 to 2006

	Number of samples	Rate of decline in hair As concentration (%)				
		Mean	Standard error	Median	Minimum	Maximum
Age						
0–9	2	35	6	35	29	40
10–19	8	47	9	53	11	77
20–29	3	62	12	53	46	86
30–39	5	63	15	71	10	95
40–49	4	61	6	59	51	76
50–59	2	47	18	47	29	65
> 60	2	29	9	29	20	37
Sex						
Female	13	51	7	51	10	95
Male	13	52	7	60	11	81
Occupation						
Non-peasants	16	48	5	46	11	81
Peasants	10	58	8	62	10	95
Total	26	52	5	53	10	95

Table 5 Comparison of hair As concentrations in different countries

Country	Number of samples	Hair As (mg/kg)		Direct As exposure	Reference
		Mean	Range		
Brazil	512	0.2	0.063–1.94	No	de Figueiredo et al., 2007
China	2134	0.73	0.43–1.03	No	Qin, 2004
Cambodia	40	1.41	0.1–7.95	Yes	Gault et al., 2008
Egypt	100	0.30 (male)		No	Saad and Hassanien, 2001
		0.29 (female)		No	
India (West Bengal)	326	6.94	1.18–31.0	Yes	Das et al., 1996
Italy	263	0.09	0.02–0.93	No	Senofonte et al., 2000
Japan	100	0.075	0.008–0.329	No	Yamato, 1988
Japan	250	0.23	< 0.72	No	Sera et al., 2002
Pakistan	187	0.43 (male)		No	Kazi et al., 2009
		0.58 (female)		No	
Sweden	114	0.085	0.034–0.319	No	Rodushkin and Axelsson, 2000
United Kingdom	1250	0.81		No	Hamilton, 1964

WHO, 1983). Qin (2004) suggested that the upper limit for As in natural hair be set to 1.0 mg/kg in healthy human subjects. This is equal to the average level of As in human hair plus a standard difference in China. In consideration of this data we set the study threshold standard at 1 mg/kg, based on which we calculated the over-standard percentage (Tables 1, 2 and 3).

3.2 Changes in hair As concentration according to occupation and age

In the human body As is transferred from blood to the hair root and deposited by binding to sulfhydryl groups of keratin, followed by movement into hair shafts (Hindmarsh, 2002; Vermeulen et al., 2009). The amount of As in longitudinal hair shaft segments reflects the As burden at the time when hair was formed (Saad and Hassanien, 2001). Thus, the total amount of As in a scalp hair sample has long been utilized as a biomarker of As exposure (Brown, 1998; Chen et al., 2005; Lin et al., 1998; Mandal et al., 2003).

Prior research in groups of people with different levels of As exposure has shown hair As concentration is significantly correlated to the degree of As exposure and indicates the level of environmental pollution (Warren, 1972). In 2002, peasant hair As concentrations were significantly higher than those of non-peasants in the study area (p = 0.004), while in 2006 there was no significant difference. In agreement with our finding, Lin et al. (2001) found the hair As concentrations for peasants who worked in fields in the contaminated area to be higher than others. The lack of significant difference between peasant and non-peasant hair As concentrations in 2006 may be due to the change in land use in study area between 2002 and 2006. When the survey started in 2002, the polluted land was still farmed and peasants were frequently in close contact with the polluted fields. Consequently, the chance of As intake was greater for peasants than non-peasants, resulting in a greater As accumulation. Between 2002 and 2006, the polluted fields were gradually abandoned, with peasants moving their agricultural activities far from the contaminated areas. The subsequent lack of contact with contaminated areas resulted in a gradual reduction in accumulated As concentrations to levels similar to non-peasants.

The survey data in 2002 showed that female hair As concentration was significantly higher than males (Table 2). Conversely, 2006 data showed that male hair As concentration was significantly higher than females (Table 2). Generally, in both non-contaminated regions and sewage irrigation areas, hair As concentration of males is higher than that of females (Hinwood et al., 2003; Wolfsperger et al., 1994). Therefore, the results for 2002 were inconsistent with prior research. This may be related to professional disparities in the study area, with females working in the field and males in manufacturing distantly from the local area. In the 2002 and 2006 samples, the proportion of female residents classified as peasants was as high as 92.3% and 80.6%, respectively. As discussed earlier, in 2002 the As concentration in the hair of peasants was significantly higher than non-peasants, while in 2006 there was no significant difference. Consequently, the As concentration differences observed between genders in the 2002 samples may arise from the females involvement in agricultural activities.

It has been shown that hair As concentrations in children are typically higher than in other age groups (Armienta et al., 1997); this was also observed in this study (Table 1). In the 2002 samples the average was higher for children, while in the 2006 samples the As concentration was significantly higher than that of other age groups. Significantly high hair As concentrations were also observed in some of the older age groups, such as the 50–59 year and over 60 year groups in 2006. Among the different age groups, hair As concentration appears to follow a trend of higher concentrations at both age extremes and lower concentrations in the middle; that is, people aged 20–29 year present the lowest concentrations and those aged 0–9 year and over 60 year the highest. This trend could be caused by different rates of As metabolism resulting in differing accumulation of As intake (Brima et al., 2006; Lokuge et al., 2004; Park et al., 2002). Following a pollution incident, the level of As accumulated in the human body is related to the strength of the metabolism (Mitra et al., 2004). Younger adults have a more robust metabolism, and therefore less As accumulates. The metabolisms of children and the elderly are correspondingly weaker, and therefore under the same exposure they may accumulate more As than other age groups.

3.3 Changes in hair As concentrations with ingestion through the food chain

Local residents immediately stopped consuming crops from the contaminated lands after the pollution occurred in the case area. They also began to obtain water via pipeline from distant mountain springs instead of from local wells. The As concentrations in their purchased agricultural products and spring water were lower than the corresponding standard safety thresholds (Table 6). Therefore, it is assumed that As consumed via the food chain was lowered to the level of non-contaminated areas.

There were eight babies in total born between 2002 and 2006 and all were included in the 2006 survey. The hair As concentrations for these children ranged from 0.93 to 6.12 mg/kg, with seven of them (87.5%) over the 1 mg/kg standard. These concentrations are extremely high despite the lack of direct exposure to contaminated food crops or drinking water, suggesting that As can also enter the human body from direct contact, air particulates and other routes. This indicates that removal of As sources from the food chain and drinking water alone is not effective in protecting local residents' health. Environmental remediation is also required to reduce land As concentrations to a safe level in order to guarantee the health of surrounding residents.

Table 6 Arsenic concentration of crops and drinking water consumed daily by Dengjiatang residents after the pollution incident reported in 2001

Sample	As conc. (mg/kg)		Standard limit (mg/kg)	Standard
	In 2006*	In 2002**		
Spinach	0.03	1.095	0.05	GB 2762-2005
Chinese cabbage	0.04	0.394	0.05	
Radish	0.01	0.175	0.05	
Celery	0.03	0.591	0.05	
Pakchoi	0.03	1.007	0.05	
Crown daisy	0.02	2.387	0.05	
Rice	0.14	–	0.15	GB 2715-2005
Water	0.0007	–	0.05	GB 5749-1985

* Data are from our study team (not published); ** data are cited from Cai et al., 2004.

4 Conclusions

Hair As concentrations in people from the As polluted area were related to gender, age and occupation (peasants or non-peasants). During the acute poisoning period, peasants and individuals who were in direct contact with contaminated soil had high concentrations of As in their hair, with no significant difference between different age groups. Four years later, differences were observed between age groups, that children and the elderly have higher hair As concentrations compared to young and middle aged individuals. The mean hair As concentration of people living in the contaminated area decreased significantly from 2002 to 2006, this could mainly be attributed to a decreased proportion of the population having high As concentrations. However, there was no obvious change in hair As concentration in people who had relatively low concentrations four years prior. Furthermore, increases were even observed in hair As concentrations for one group of the population. Therefore, As pollution has a long-term continuous influence on the health of local residents.

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