

ELEVATED LEVELS OF ARSENIC AND FLUORIDE, BUT NOT SELENIUM, ASSOCIATED WITH ENDEMIC DISEASE IN THE CHINESE VILLAGE OF DAZHUYUAN, SHAANXI PROVINCE

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SUMMARY: In this investigation, we determined the concentrations of arsenic (As), fluoride (F), and selenium (Se) in the environment (coal, drinking water, soil, corn, and chili) and in biological samples from local residents (urine, hair, and serum) from the endemic disease village of Dazhuyuan and the non-endemic disease control village of Liu in Shaanxi Province, China. Except for drinking water, the As and F levels of all samples from Dazhuyuan were significantly higher than those of the control. In contrast, although the Se contents in the environment of Dazhuyuan village were remarkably greater than those of the control, the Se levels in tissue samples from the exposed and control populations were similar. This study found for the first time that naturally occurring, high levels of As, F, and Se in the Dazhuyuan environment could contribute to local residents suffering from arsenicosis and fluorosis but not selenosis.

Keywords: Arsenic pollution; Dazhuyuan, Shaanxi Province; Fluoride pollution; Food contamination; Selenium pollution.

INTRODUCTION

Southern Shaanxi Province is one of the regions in China with severe endemic diseases. Previous studies reported endemic arsenicosis due to coal combustion or drinking water,^{1,2} endemic fluorosis due to coal usage or to drinking water,^{2,3} and selenosis due to coal burning.⁴ However, each of these separate reports focused exclusively on one kind of chronic endemic poisoning. Surprisingly, there are no previous reports on the combined elevated levels of arsenic (As), fluoride (F), and selenium (Se) as environmental contaminants or their resulting levels in tissues and fluids of exposed individuals. Since environmental contaminants can have additive, antagonistic, or synergistic effects, we initiated the present investigation in the Dazhuyuan (an exposed area) and Liu (a non-exposed area) villages of Shaanxi Province as a preliminary survey focusing on As, F, and Se levels in the environment and in the population.

MATERIALS AND METHODS

Investigated areas and patient diagnostic criteria: This investigation was initiated on October 25, 2005. Two villages (Dazhuyuan and Liu), approximately 260 km apart, were selected for study. Dazhuyuan village, in the southeastern part of Shaanxi Province, is located in an endemic disease area, whereas Liu village, situated 25 km south of Xi'an city, is a non-endemic disease area. Residents were randomly selected from Dazhuyuan (198 residents) and Liu (88 residents). Patients affected by As toxicity were diagnosed based upon National Standard of

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Diagnosis for Endemic Arsenicosis in China (WS/T 211–2001). Skeletal fluorosis was defined according to the standard of Chinese Health Ministry (WS 192-1999), and selenosis was diagnosed by the loss of hair and nails. Written informed consent was obtained from the participants, and the ethics committee of Xi'an Jiaotong University approved the design of the study.

Sample collection and storage: In Dazhuyuan village, samples were collected from five different locations (east, west, south, north, and the centre). These included 10 coal samples, 20 drinking water samples, 20 soil samples, 20 corn, and 20 chili samples, all of which were collected from the participants' homes. Drinking water and spot urine samples were collected in 25-mL polypropylene tubes and immediately refrigerated until analyzed without delay at our university laboratory. Approximately 0.5 g of new growth hair was cut and sealed in envelopes at room temperature, as were coal, corn, and chili samples. Blood samples were immediately centrifuged and the resulting serum was refrigerated in polypropylene tubes and analyzed within one week. All the samples from the control site were collected in identical manner and stored in the same manner.

Determination of As, Se, and F levels: The concentrations of As and Se were determined using a graphite furnace atomic absorption spectrophotometer (Hitachi 180–80 Polarized Zeeman Atomic Absorption Spectrophotometer, Tokyo, Japan). The F levels in samples were determined using an F ion selective electrode (Shanghai Exactitude Instrument Company, China).

Statistical analysis: All data are presented as mean values with standard deviations (SD). Statistical tests for significance were performed using the Student t test.

RESULTS

The demographic distributions of these two villages were similar (Table 1). In all samples but drinking water, the As and F levels were significantly greater in Dazhuyuan than those of the control village, Liu (Tables 2 and 3). The Se levels were significantly elevated in each environmental source from Dazhuyuan compared to Liu (Table 2). However, Se levels in biological samples were similar among subjects from both villages (Table 3).

Table 1. Characteristics of subjects in the two villages

Characteristics	Dazhuyuan village		Liu village	
	Male (n=100)	Female (n=98)	Male (n=45)	Female (n=43)
Mean age (SD) (yr)	48.0 (19.1)	48.2 (17.8)	46.1 (13.7)	49.7 (11.1)
Age range (yr)	11-83	11-83	18-75	22-74
≤ 30 yr	19	15	10	8
31–50 yr	32	37	13	15
> 51 yr	49	46	22	20
Period of resident (SD) (yr)	46.9 (18.9)	39.6 (20.1)	45.9 (15.2)	38.9 (16.3)
Arsenicosis (%)	37 (37.0)	23 (23.5)	0 (0)	0 (0)
Skeletal fluorosis (%)	6 (6.0)	4 (4.1)	0 (0)	0 (0)
Selenosis (%)	0 (0)	0 (0)	0 (0)	0 (0)

Table 2. Levels of As, F, and Se in environmental samples from the two villages

Elements in samples	Dazhuyuan village		Liu village		t value	p value
	n	Mean ± SD	n	Mean ± SD		
As in coal (mg/kg)	10	109.16 ± 16.53	10	1.38 ± 0.34	20.62	0.00*
F in coal (mg/kg)	10	1757.46 ± 634.61	8	120.29 ± 118.51	7.16	0.00*
Se in coal (mg/kg)	10	2.37 ± 0.24	10	0.14 ± 0.11	27.26	0.00*
As in drinking water (mg/L)	20	0.007 ± 0.002	16	0.008 ± 0.002	-0.82	0.42
F in drinking water (mg/L)	20	0.15 ± 0.16	16	0.12 ± 0.02	0.97	0.35
Se in drinking water (mg/L)	20	0.015 ± 0.004	16	0.011 ± 0.002	3.36	0.00*
Soluble As in soil (mg/kg)	20	0.72 ± 0.27	20	0.46 ± 0.12	3.76	0.00*
Soluble F in soil (mg/kg)	20	1.47 ± 1.25	20	0.64 ± 0.62	2.65	0.01*
Soluble Se in soil (mg/kg)	20	0.02 ± 0.01	20	0.01 ± 0.002	9.51	0.00*
As in corn (mg/kg)	20	0.87 ± 0.65	15	0.34 ± 0.15	3.51	0.00*
F in corn (mg/kg)	20	4.78 ± 1.56	20	2.89 ± 1.39	4.01	0.00*
Se in corn (mg/kg)	20	2.97 ± 1.59	15	1.30 ± 1.02	3.78	0.00*
As in chili (mg/kg)	20	0.92 ± 0.60	15	0.34 ± 0.15	4.42	0.00*
F in chili (mg/kg)	20	31.79 ± 21.75	15	7.98 ± 2.90	4.60	0.00*
Se in chili (mg/kg)	20	1.34 ± 0.98	15	0.78 ± 0.45	2.36	0.02*

Compared with the control area, *p<0.05.

Table 3. Levels of As, F, and Se in tissues and fluids from subjects in each of the two villages

Elements in samples	Dazhuyuan village		Liu village		t value	p value
	n	Mean ± SD	n	Mean ± SD		
Urinary As (mg/L)	72	0.04 ± 0.02	44	0.03 ± 0.01	5.00	0.00*
Urinary F (mg/L)	72	2.70 ± 1.93	44	0.55 ± 0.33	9.22	0.00*
Urinary Se (mg/L)	72	0.03 ± 0.01	44	0.03 ± 0.01	-0.07	0.95
Hair As (mg/kg)	85	4.84 ± 1.88	49	0.52 ± 0.91	20.90	0.00*
Hair F (mg/kg)	56	24.47 ± 21.77	42	8.80 ± 6.55	5.09	0.00*
Hair Se (mg/kg)	85	0.50 ± 0.13	49	0.49 ± 0.10	0.75	0.46
Serum As (mg/L)	91	0.05 ± 0.03	40	0.03 ± 0.01	5.12	0.00*
Serum F (mg/L)	91	0.31 ± 0.19	40	0.03 ± 0.03	14.13	0.00*
Serum Se (mg/L)	91	0.10 ± 0.05	40	0.09 ± 0.04	1.26	0.21

Compared with the control group, *p<0.05.

DISCUSSION

The average As level of coal in the Dazhuyuan village (109.2 mg/kg) is much higher than the average values in China (4.5 mg/kg)⁵ and the world (9.0 mg/kg).⁶ However, it was lower than As levels (2,167 mg/kg) in Bazhi, Guizhou Province, China, which is an area of endemic arsenicosis due to coal.⁷ Levels of As in corn and chili from Dazhuyuan were 4 to 18 times higher than the maximum levels in these foods in China (0.2 mg/kg and 0.05 mg/kg, respectively, GB 2762–2005). The As concentration in hair from the Dazhuyuan villagers (4.84 mg/kg) was significantly higher than previously reported average in Chinese residents (0.93 mg/kg).⁸ Besides, we determined that 60 of 198 subjects (30.3%) from Dazhuyuan had manifestations of arsenicosis whereas none of 88 subjects from Liu were affected. Based on the standard for chronic environmental As poisoning (WS/T 183–1999), these data suggest that Dazhuyuan village is an area of endemic arsenicosis.

Coal F content in Dazhuyuan (1,757.5 mg/kg) was considerably greater than the mean for Chinese coal (136 mg/kg)⁹ and much greater than the estimated safety threshold of coal F (190 mg/kg).¹⁰ The F contents in corn and chili of the exposed village surpassed by 2 to 31 times the maximum levels of contaminants in foods of China (1.5 mg/kg for corn and 1.0 mg/kg for chili, GB 2762-2005). Urinary F concentrations in Dazhuyuan (2.70 mg/L) were higher than samples from residents of Chinese cities without significant fluoride pollution (0.59 mg/L in Guangzhou, 0.58 mg/L in Chengdu, and 0.56 mg/L in Shenyang, respectively)¹¹ and higher than in Hungary (0.29 mg/L).¹² Hair F levels (24.47 mg/kg) in Dazhuyuan were higher than those in Poland without significant fluoride exposure (2.00 mg/kg)¹³ and higher than those of skeletal fluorosis patients (1.09 mg/kg) in Inner Mongolia, China where fluorosis is attributable to drinking brick tea.¹⁴ Similarly, serum F concentrations in Dazhuyuan village were higher than levels found in Jiangsu Province.¹⁵ Also, Chen et al.¹⁶ reported that the prevalence of dental fluorosis was more than 30.0% in the children in Ankang area where Dazhuyuan is located; and we identified 10 skeletal fluorosis patients in Dazhuyuan. Based on the standard for endemic fluorosis (GB 17017-1997), these data indicate that Dazhuyuan is an endemic fluorosis area.

Se levels in coal and soil from Dazhuyuan village were higher than the means in China (2 mg/kg and 0.01 mg/kg, respectively).^{17,18} Se in the drinking water of Dazhuyuan was higher than levels defined by the Chinese Drinking Water Sanitation Standard (0.01 mg/L, GB 5749-85). Se levels in corn and chili far surpassed the tolerance limits in foods of China (0.3 mg/kg and 0.1 mg/kg, respectively, GB 13105-91). By each of these measures, Dazhuyuan exceeds the acceptable environmental threshold levels of selenium.¹⁹ However, the Se levels in biological samples from individuals in Dazhuyuan did not exhibit significant increases when compared with the control region, and the levels were also close to those seen in non-Se-exposed adults in India, USA, and Taiwan.²⁰⁻²² Similar results have been reported previously by Shi et al. in a study focused on selenium levels in the same area.²³ The discrepancy might be due to the high arsenic levels in the body since arsenic can markedly increase the excretion of Se²⁴ and thus serve to alleviate Se poisoning.²⁵ Consistent with this observation, no selenosis patients were identified in Dazhuyuan (Table 1).

In conclusion, all available data showed that Dazhuyuan has high environmental levels of As, F, and Se. Local residents also suffer from elevated rates of arsenicosis and fluorosis but not selenosis.

Presentation of this work was given at the XXVIIIth Conference of the International Society for Fluoride Research in Toronto, Ontario, Canada, 7–11 August 2008.

ACKNOWLEDGEMENTS

This work was supported by a grant from the Department of Controlling Endemic Diseases, Shaanxi Province, China (2005-2007). We thank Professor Peter J. Stambrook, University of Cincinnati College of Medicine, for critical reading of the manuscript. The authors are also grateful to ScienceDocs, Inc. for their editing of a previous manuscript.

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