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Fluoride in drinking water, brick tea infusion and human urine in two counties in Inner Mongolia, China

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ABSTRACT

The objective of this study was to detect the fluoride level in the drinking water and the urine of habitants aged 16–55 years living in Inner Mongolia China. Furthermore, fluoride concentration of the brick tea infusion samples which were drunk by Mongolia herdsmen in everyday life living in SumuErga village of Ejin Horo Banner, Inner Mongolia China was also determined. A total of 117 participants (61 female and 56 male) were recruited from two counties for a cross-sectional study on health effects of chronic fluoride exposure from drinking water and drinking brick tea infusion. The fluoride concentration in drinking water, urine and brick tea infusion samples were determined using fluoride ion selective electrode method obtained from the Ministry of Health of the People's Republic of China. The average fluoride concentration in drinking water samples was 0.32 ± 0.01 mg/L at AretengXire town of Ejin Horo Banner, 0.70 ± 0.19 mg/L at SumuErga village of Ejin Horo Banner, and 2.68 ± 1.15 mg/L at ZhalaiNuoer district of Manzhouli city. The average fluoride concentration in brick tea infusion samples which collected from Mongolia herdsmen at SumuErga village of Ejin Horo Banner was 1.81 ± 1.09 mg/L. The average urinary fluoride concentration at AretengXire town of Ejin Horo Banner was 0.59 ± 0.48 mg/L, at SumuErga village of Ejin Horo Banner was 1.45 ± 0.93 mg/L and at ZhalaiNuoer district of Manzhouli city was 3.06 ± 1.53 mg/L. The higher fluoride levels in the urine of participants may be associated to higher fluoride in drinking water at ZhalaiNuoer of Manzhouli city. However, drinking brick tea infusions with higher fluoride may be the cause of the higher fluoride contents in the Mongolia herdsmen's urine.

1. Introduction

Fluoride is a necessary element to human health, and a moderate amount of fluoride intake is confirmed that it is the effective way of reducing dental caries among children and adult [1]. However, excessive fluoride intake through drinking water or food results in dental fluorosis and skeletal fluorosis [2], furthermore, non-skeletal phase damage such as parathyroid [3], kidney and liver [4] is observed.

Endemic fluorosis is widely distributed throughout the world, covering over forty countries such as India, Mexico and Africa etc. [5–8]. It is also extensively distributed in China except Shanghai and Hainan province, and more than 1.34 million patients who are distributed in 1115 counties, has suffered from skeletal fluorosis due to high fluoride drinking water in 2006 [9]. In China, however, in addition to cases of endemic fluorosis from drinking water, there are other two types of endemic fluorosis, which are caused by the pollution from coal burning and by drinking brick tea with high fluoride concentration [10]. It has been found that drinking brick tea type fluorosis is prevalent mainly among the minority inhabitants in the west of China [10], and the prevalence of fluorosis provoked by brick tea drinking has attracted more and more attention [11–13].

The main pathway of fluoride excretion was via the kidney, and urinary fluoride excretion corresponded to about 50% of fluoride intake [14]. Many studies found that urinary fluoride concentration was correlated to the amount of fluoride intake, and then fluoride content in urine is considered as a good indicator for reflecting fluoride exposure [15].

Inner Mongolia lies in north-west of China. More than two million Mongolian herdsmen are living in there. They are favor of drinking brick tea in daily life, and each adult herdsman can consume about 2 L brick tea infusions. However, the Han habitants who live in Inner Mongolia seldom drink brick tea infusion. Drinking brick tea has been a habit for a long time, but little information of fluoride levels in brick tea infusions and urine of herdsmen living in Inner Mongolia is available, especially in middle-west area of

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Inner Mongolia. The purpose of this investigation was to measure and compare the urinary fluoride levels in adults in the study of the health effects of chronic fluoride exposure from drinking water or drinking brick tea infusions. We used an ion selective electrode method to determine fluoride in drinking water, brick tea infusions and urine samples. We are reporting fluoride levels in drinking water from three sites of two counties in Inner Mongolia as well as urinary fluoride from participants from each site. Additionally, fluoride levels in brick tea infusion of one village are also reported.

2. Materials and methods

2.1. Site description and subject selection

In 2005–2006, a cross-sectional study was conducted in two counties (Ejin Horo Banner and Manzhouli city) in north-west of China, located in Inner Mongolia (Fig. 1).

Ejin Horo Banner, lies between longitudes 108°58′ and 110°25′ and between latitudes 38°56′ and 39°49′. The climate of this area is arid with mean annual temperature 6.2 °C and annual precipitation 358.2 mm, altitude between 1070 and 1556 m. The total area of this Banner is 5899.6 km^2, and the average population density is 23 person/km^2. Another study area, Manzhouli city is a smaller city of the Inner Mongolia, China, located between longitudes 117°12′ and 117°53′ and latitude 49°19′. The climate of this area is semi-arid with mean annual temperature 1.3 °C and annual precipitation 299.5 mm, altitude between 500 and 1000 m. The total geographical area of the city is 696.3 km^2, and the average population density of this city is 196 person/km^2.

Thirty-eight Han participants (20 men and 18 women) were recruited from ZhalaiNuoer district of Manzhouli city, Inner Mongolia. Manzhouli is a city known by its high fluoride content in drinking water (around 2.0 mg/L), and dental fluorosis in children and about 40% detection rate of skeletal fluorosis in adult had ever been reported in 1999 [16]. A second group of 49 Mongolian participants accustomed to drinking brick tea were recruited from SumuErga village of Ejin Horo Banner (18 men and 31 women). A third group consisted of 30 Han participants (18 men and 12 women) seldom drinking brick tea were recruited from AretengXire town of Ejin Horo Banner where most of the habitants were drinking tap water from the same waterworks.

2.2. Water samples

Except habitants living in AretengXire town of Ejin Horo Banner who drink tap water, shallow groundwater is the main water source available for the habitants of other two study sites, and it is extracted by hand pumps or motor pumps. The water samples were collected from the participant family and put into clean polyethylene bottles of 0.25 L capacity, which were stored at 4 °C until they were analyzed.

2.3. Urine samples

The stochastic urine samples were collected into the clean polyethylene bottles, transported and stored in ice bag with no more than 12 h. Then the samples were stored at −18 °C until the analysis was performed.

2.4. Brick tea infusion samples

Brick tea infusion samples were collected in clean polyethylene bottles and were stored at −18 °C until they were analyzed. All of the brick tea infusion samples were from the Mongolian herdsmen family living in SumuErga village of Ejin Horo Banner.

2.5. Analytical methods

2.5.1. Fluoride in water

The fluoride concentration in drinking water was determined electrochemically, using the Ministry of Health of the People's Republic of China ion selective method. This method is applicable to the measurement of fluoride in drinking water, groundwater and lightly polluted water in the concentration range of 0.2–2000 mg/L [17]. The fluoride concentration in the water sample was determined directly after diluting with equal volume of total ionic strength adjustment buffer (TISAB) of pH 5.2, and the detailed composition is listed in Table 1. The electrode used was a CSB-F-1 fluoride electrode (Changsha Manufactory of Semiconductor,

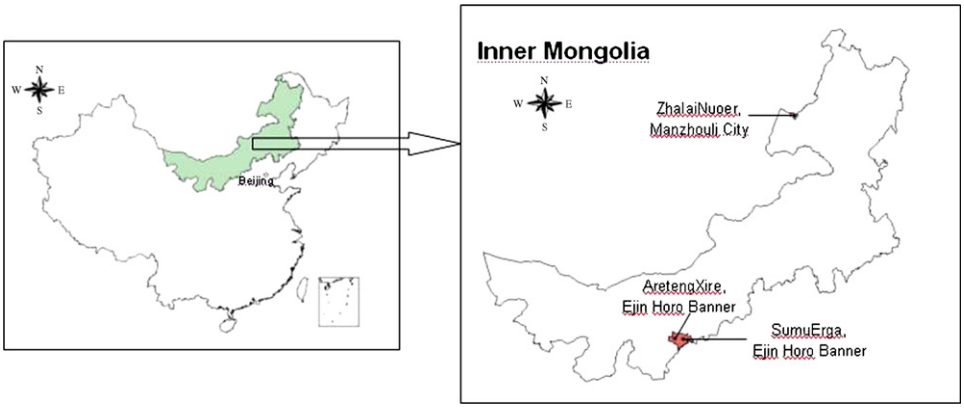

Fig. 1. Location map of the study area.

Table 1
Different compositions of TISAB solution for determination fluoride in water, urine and brick tea infusion samples.

	Water	Urine	Brick tea infusion
Compositions of TISAB solution	58 g NaCl; 10 g $Na_3C_6H_5O_7{\cdot}2H_2O$; 57 mL glacial acetic	58 g NaCl; 4 g $Na_3C_6H_5O_7{\cdot}2H_2O$; 57 mL glacial acetic	58 g NaCl; 120 g $Na_3C_6H_5O_7{\cdot}2H_2O$; 57 mL glacial acetic
Method to adjust pH to 5.2	5 mol/L NaOH	5 mol/L NaOH	5 mol/L NaOH
Total volume	1 L	1 L	1 L

China), coupled to an Orion 868 electrometer. Standards solutions (0.2–10 mg/L) were prepared from a stock solution (100 mg/L) of sodium fluoride.

2.5.2. Fluoride in urine

In order to analyze fluoride in urine, the Ministry of Health of the People's Republic of China method for the determination of fluoride in urine was used, and the Method Quantitation Limit of detection for fluoride in urine was 0.05 mg/L [18]. The fluoride concentration in the urine sample was determined directly after diluting with equal volume of TISAB of pH 5.2, and the detailed composition is listed in Table 1. Then, fluoride was quantified using a fluoride ion specific electrode Orion 868 Electrode, as previously described.

2.5.3. Fluoride in brick tea infusion

To determine fluoride in brick tea infusion, the method reported by Yu [19] was used. The fluoride concentration in the brick tea infusion sample was determined directly after diluting with equal volumes of TISAB of pH 5.2, the detailed composition is listed in Table 1. Then, fluoride was quantified using a fluoride ion specific electrode Orion 868 Electrode, as previously described. The Method Quantitation Limit of detection for fluoride in brick tea infusion was 0.2 mg/L.

All the reagents were of analytical-reagent grade or better. The purified water for all dilutions was of 18.3 MΩ cm^{-1} purity using the Milli-Q (Millipore, Bedford, MA, USA) deionization system.

2.5.4. Quality control

The accuracy of measurements was assessed with addition method in water, urine, and brick tea infusion samples [20]. Recoveries of addition samples were 96.19% from water, 96.47% from urine, and 96.70% from brick tea infusion. The method repeatability was tested by analyzing seven replicates of one water sample, one urine sample and one brick tea infusion sample separately. The coefficients of variation in the water, urine and brick tea infusion sample replicates were 2.15% (mean = 0.41 mg/L, $n = 7$), 3.59% (mean = 0.58 mg/L, $n = 7$), and 1.88% (mean = 3.28 mg/L, $n = 7$) respectively.

2.6. Statistical analysis

Statistical analyses were carried out using STATISTICA® (Version 5.0) for Windows (StatSoft, Inc., USA, 1995).

3. Results and discussion

Table 2 shows the fluoride levels in drinking water in the three study sites. The mean fluoride concentration was 0.70 ± 0.19 mg/L at SumuErga village of Ejin Horo Banner, 0.32 ± 0.01 mg/L at AretengXire town of Ejin Horo Banner, and 2.68 ± 1.15 mg/L at ZhalaiNuoer district of Manzhouli city. China National Standard for Fluoride in Drinking Water is 1.0 mg/L. In the present study, except for one water sample which exceeded the permissible limit a little bit, the fluoride concentration in drinking water was at normal range at Ejin Horo Banner. Contrarily, 92% of the samples exceeded the maximum permissible limit at ZhalaiNuoer district of Manzhouli city. Generally, the adult daily water consumption is considered as 2.5 L, if we take average fluoride concentration in the different water sources into consideration then the fluoride intake by adult from ingested water could be 0.80, 1.75 and 6.70 mg at AretengXire in Ejin Horo Banner, SumuErga in Ejin Horo Banner and ZhalaiNuoer in Manzhouli city, respectively. The dietary fluoride intake allowance is 3.50 mg/(person · day) for adults. It is assured from the results that the people at ZhalaiNuoer in Manzhouli city are chronically exposed to higher fluoride levels from drinking water.

Table 2
Fluoride concentration in drinking water samples in the study area (mg/L).

Locality	Mean	±S.D.	Min	Max	*n*
AretengXire, Ejin Horo Banner	0.32	0.01	0.31	0.33	2
SumuErga, Ejin Horo Banner	0.70	0.19	0.40	1.13	14
ZhalaiNuoer, Manzhouli City	2.68	1.15	0.52	4.10	26

n means the sample numbers.

Table 3
Fluoride concentration in brick tea infusions in the study area (mg/L).

Locality	Mean	±S.D.	Min	Max	*n*
SumuErga, Ejin Horo Banner	1.81	1.09	0.77	6.06	39

n means the sample numbers.

Table 3 displays the fluoride content of the brick tea infusions drinking by Mongolian herdsmen living in SumuErga, Ejin Horo Banner. Brick tea infusions had a fluoride concentration varying from 0.77 to 6.06 mg/L, 82% of the brick tea infusion samples exceeded the maximum permissible limit of fluoride in drinking water and the highest fluoride concentration in the brick tea infusions was 6.06 mg/L, which was five times higher than the accepted level of China National Standards for Fluoride in Drinking Water.

We observed that the brick tea infusions consumption was approximately 2.16 L/(adult · day) in SumuErga, Ejin Horo Banner, if we take average fluoride concentration in the brick tea infusions into consideration then the fluoride intake by adult from ingested brick tea infusion could be 3.90 mg in SumuErga, Ejin Horo Banner. Its value is higher than the China Hygienic Standard for Daily Total Intake of Fluoride, 3.5 mg. It is evident from the result that Mongolia herdsmen who live at SumuErga, Ejin Horo Banner are chronically exposed to higher level of fluoride from drinking brick tea infusion.

The persons, who are not exposed to excessive levels of fluoride, the fluoride concentration in their urine is usually in the range of 1.0–1.5 mg/L [21]. Urinary fluoride concentrations in adult participants from AretengXire of Ejin Horo Banner, SumuErga of Ejin Horo Banner and ZhalaiNuoer of Manzhouli city are displayed in Table 4. The urinary fluoride content of adult from all the three habitations had significant difference. The fluoride content of urine samples collected from AretengXire of Ejin Horo Banner ranged from 0.06 to 2.06 mg/L, and 94% of the fluoride content in the urine samples was lower than 1.5 mg/L, and some habitants with very slight dental fluorosis and two cases with slight skeletal fluorosis were observed in AretengXire. At SumuErga of Ejin Horo Banner, the urinary fluoride concentration varied from 0.14 to 4.30 mg/L, and 34% of the urinary fluoride concentration was higher than 1.5 mg/L. The investigated herdsmen had 75% dental fluorosis prevalence rate and 31% skeletal fluorosis detection rate at SumuErga. The highest urinary fluoride content appeared in the habitants living at ZhalaiNuoer of Manzhouli city, ranged from 0.78 to 7.30 mg/L, and 87% of the fluoride content in urine sample was higher than 1.5 mg/L. The adult dental fluorosis prevalence rate was 72% and skeletal fluorosis detection rate was 63% of the habitants at ZhalaiNuoer, which was higher than those reported by Zhou et al. in 1999 [16].

Additionally, as shown in Fig. 2, urinary fluoride concentration for males did not exhibit a difference when compared to the

Table 4
Urinary fluoride concentration in the study area (mg/L).

Locality	Mean	±S.D.	Min	Max	*n*
AretengXire, Ejin Horo Banner	0.59	0.48	0.06	2.06	30
SumuErga, Ejin Horo Banner	1.45	0.93	0.14	4.30	49
ZhalaiNuoer, Manzhouli City	3.06	1.53	0.78	7.30	38

n means the sample numbers.

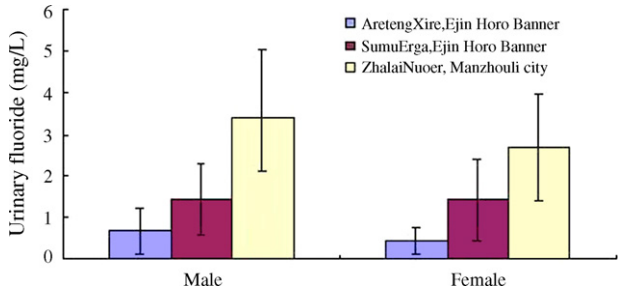

Fig. 2. Sex dependent urinary fluoride concentration in adult in study area (mg/L). Error bars represent ±S.D.

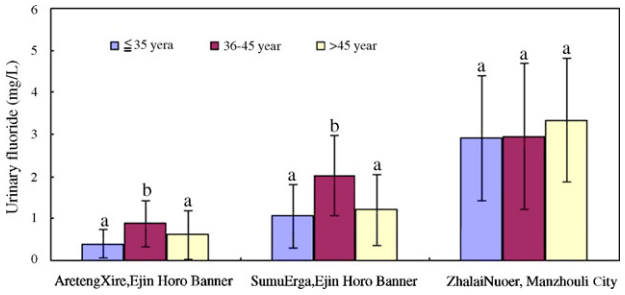

Fig. 3. Age dependent urinary fluoride concentration in adult in study area (mg/L). Error bars represent ±S.D. The data with different superscript letters are significantly different from each other ($P < 0.05$) according to General Univariate ANOVA test.

urinary fluoride concentration for females. On the univariate analysis, urinary fluoride level significantly differed in different age groups in which the highest fluoride level was in the 36–45 years group, and the lowest level was in the age group of ≤35 years at AretengXire and SumuErga of Ejin Horo Banner. However, urinary fluoride level was not significantly associated with age at ZhalaiNuoer of Manzhouli city (Fig. 3).

4. Conclusions

Urinary fluoride concentration is a good indicator of fluoride exposure in a population. In the study, two kinds of fluoride excess intake were reported. Although fluoride level in drinking water was lower than 1.0 mg/L, the higher fluoride levels in brick tea infusions caused the high fluoride contents in the urine of Mongolia herdsmen who were accustomed to drinking brick tea infusions in everyday life, and some adult herdsmen had been affected by dental fluorosis and skeletal fluorosis at SumuErga of Ejin Horo Banner. Additionally, 92% of drinking water samples had higher fluoride concentration (>1.0 mg/L) at ZhalaiNuoer of Manzhouli city, and the higher fluoride levels of adult urine at this area were linked to higher fluoride content in drinking water, and many habitants suffered from fluorosis. In Inner Mongolia of China, brick tea and drinking water with high fluoride content are the two pathways for fluorosis. The drinking high fluoride water fluorosis has attracted close attentions for a long time, unfortunately, most of Mongolian herdsmen who drink brick tea infusions as daily beverage are unaware of the health risk of the large amount of fluoride being ingested, therefore, it is urgent to examine the fluoride exposure to brick tea and its association with long-term health effects as well as searching for manufacturing the brick tea with lower fluoride.

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