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# Fluorine distribution in aquatic environment and its health effect in the Western Region of the Songnen Plain, Northeast China

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**Abstract** Endemic fluorosis was investigated and studied in the west region of the Songnen plain, Northeast China in 2001–2002. The results showed that the fluorine distribution in aquatic environment was that the fluorine concentrations in the lake water and unconfined ground water were higher than that in the river water and confined ground water. The lake water (Alkali lake) is connected with unconfined ground water. In unconfined ground water, from the east and southeast areas to the west and the northwest areas of the plain, fluorine concentration fluctuated with high and low alternatively. The fluorine in the water comes from the weathering of rocks and minerals in the mountains and hills around the Songnen Plain. The main influence factors of the fluorine distribution in aquatic environment are discussed. Unconfined ground water containing high fluorine is used as drinking water. In this region, the fluorine concentration in drinking water is evidently correlated to the morbidity of dental and skeletal fluorosis. High fluorine concentration in drinking water has endangered human health.

**Keywords** Fluorine · Aquatic environment · Unconfined ground water · Drinking water · Songnen plain

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## Introduction

Fluorine is one of important life elements to human health. Low levels of fluorides from drinking water or food can help presenting dental cavities. At high levels, fluorides can result in tooth and bone damage, especially to children. Much attentions have been paid on fluorine in the worldwide (Cao et al. 2000; Carton 2006; Subba and Devadas 2003). It was reported that high fluorine concentration in unconfined ground water which was consumed as drinking water by local residents has resulted in the dental fluorosis and the skeletal deformation fluorosis and strongly risk to human health in the western plain of Jilin province and Heilongjiang province, Northeast China, (Li and Xu 1987; Meng et al. 1987). It is one of endemic fluorosis area that caused by drinking water that containing the high fluorine in China (Tan 1991). Endemic fluorosis has been effecting and threatening on human health in this region. And the sustainable development of the economy has been limited. This paper attempts to describe the fluorine distribution, explain

influence factors and health effects of the fluorine in water environment, including the surface water and ground water.

## Study site

The Songnen Plain is located in the middle and west part of the Northeast, China (Fig. 1), which includes the western parts of the Jilin province and the Heilongjiang province. Geographical location is 42°31′~51°20′ N, 121° 40′~128°30′ E (Liu 2001). This plain was mainly formed by the fluvial and the alluvial accumulations of the Songhuajiang River system and the Nenjiang River system. The East, North and West parts of the plain are bordered on the Changbai mountains, the Xiaoxinganling mountains and the Daxingan mountains, respectively. The southern part is bordered on the Liaohe River watershed. The total area of this plain is $17.8\times10^4$ km$^2$. The Nenjiang River runs from the northwest to the middle part of the plain, and enters in the Songhuajiang River, then runs into the northeast part in the Heilongjiang Province. The topography is higher in the northwest and southeast parts, and lower in the west and southwest parts of this region. This typical subsided basin was formed in the Mesozoic era and Cenozic era. This region is mainly underlain by the stratums of the cretaceous, quaternary and tertiary ages. It is a typical vulnerable zone of eco-environment in the western part of the Songnen plain, Northeast China. The climate is from the humid and semi-humid in the east and the southeast parts to the semi-arid and arid climate in the west and northwest parts and both are affected by continental monsoon in this plain.

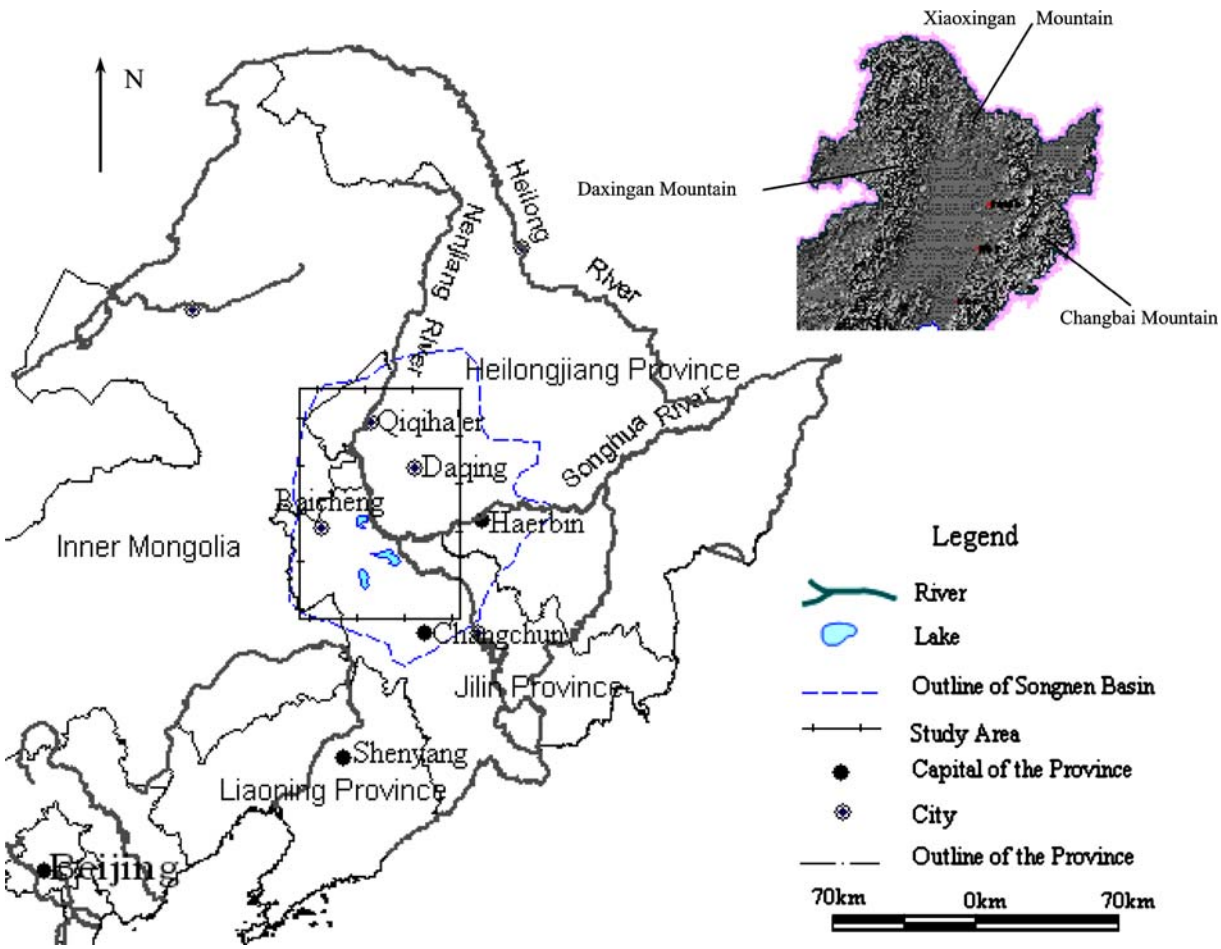


**Fig. 1** Study region location in the Songnen plain of Northeast China

Annual precipitation is about 350~500 mm, and annual average temperature is about 0~5°C. There are a few of aquifers in the ground water of this region (Liao and Lin 2004). The main aquifers include unconfined ground water and confined ground water. Unconfined ground water is main water source that is often used as drinking water by local residents. Soil type includes phaeozem–cherozem–chestnut soil from the east and the southeast parts to the west and the northwest part (Institute of Forest—Soil of Chinese Academic Sciences 1980). Also, vegetation type is the forest steppe-steppe-arid steppe from the east and the southeast parts to the west and the northwest part.

## Materials and methods

### Sample collection

Water samples were collected in 2001–2002, which included surface water (river water and lake water) and ground water (unconfined ground water and confined ground water). In general, ground water samples were collected from the Baicheng, the Songyuan regions in the Jilin province, and from Qiqiaer, Nenjiang, Daqing, Zhaozhou, Zhaoyuan and Zhaodong regions in the Heilongjiang province. Total of 1,100 water samples were evenly distributed in the total 25 counties in administration. The fluorine concentration in the water samples was determined with ion selective electrode method in the sites. Other elements in the water samples were determined with normal methods in the laboratory. At the same time, endemic fluorosis (dental fluorosis and skeletal fluorosis ) in the study region was investigated. Data and literatures on the hydrochemistry and endemic fluorosis have been collected during the investigation (Office of Leader Group of Control Endemic Disease in Jilin province 1996; Jiao 1994).

### Data processing

Data in hydrochemistry has been processed by Grapher, Surfer, SPSS10.0, and ArcGIS 9.0.(Zhang et al. 2001).

## Results

### Fluorine concentration in the surface water

There are Songhuajiang river system and Nenjiang river system in the Songnen plain region. In general, the fluorine concentration in the surface water was lower than that in the ground water. This is consistent with Zhang Xue-lin's results (Zhang et al. 1996). The fluorine concentration in the surface water was lower due to less contact with rocks and showed a normal or lognormal distribution (Table 1).

The fluorine concentration in the lakes and shallow waters (bog, swamp, and so on) was higher than that in the rivers. The fluorine concentration in the lakes and shallow waters ranged from 1.05–12.2 mg/l. The highest value of the fluorine concentration in the lakes and shallow water was mainly distributed in Hua-ao lake, Dabushu lake and Dawang lake and so on. Fluorine concentration in the river waters ranged from 0.13–0.20 mg/l. The water source in alkali lakes is closely connected with the groundwater. The fluorine content in the alkali lakes has been accumulated due to the evaporation on the surface water and change of hydro-chemical features. For example, since $HCO_3^-$ concentration in the alkali lakes is relatively decreased, $CO_3^{2-}$ is easily combined with $Ca^{2+}$, $Mg^{2+}$ to form $CaCO_3$, and $MgCO_3$, and deposit in the sediments. But it is difficult to form $CaF_2$ in the sediments. At the same time, since $OH^-$ is easily combined with $Fe^{3+}$, $Al^{3+}$ to form stable $Fe(OH)_3$ and $Al(OH)_3$ in the sediments, also $Fe^{3+}$ and $Al^{3+}$ oxidations could not

**Table 1** Fluorine background levels in river water (mg/l)

| Site | Sample number | Range value | Middle value | Arithmetic mean value | | Geometric mean value | | Probability distribution type |
|---|---|---|---|---|---|---|---|---|
| | | | | (*X*) | (*S*) | (*G*) | (*Sg*) | |
| Songhuajiang river | 29 | 290.10–0.76 | 0.14 | 0.21 | 0.16 | 0.17 | 1.80 | Lognormal distribution |
| Second Songhujiang river | 11 | 110.10–0 .76 | 0.20 | 0.30 | 0.21 | 0.24 | 2.01 | Lognormal distribution |
| Nenjiang river | 13 | 130.10–0.22 | 0.13 | 0.14 | 0.04 | 0.14 | 1.32 | Normal distribution |

been combined with fluorine($F^-$). Therefore, free fluorine ions ($F^-$) in the alkali lakes are increased (Hitchon 1995). In addition, it was found that there was negative correlation in relationship between $Ca^{2+}$, $Mg^{2+}$ and $F^-$concentrations in the alkali lakes, $R_{Ca}$=−0.849, $R_{Mg}$=−0.881, $p$<0.01 (Table 2).

## Distribution of fluorine concentration in the groundwater

The contour map of fluorine concentrations and three-dimensional diagram were plotted using Surfer software based on the data of fluorine concentrations in unconfined ground water in the west part in Songnen plain region. The contour map of fluorine concentrations in unconfined ground water (Fig. 2) showed that the fluorine distribution alternated with high and low concentration intervals from the south to the north part of the plain. High and low concentration alternated trend was between the Songhuajiang river and the Liao river in the southern part of the Songnen Plain and between the Taoer River and the Nenjiang River in the northern part of the Songnen plain. There were two low fluorine zones and three high fluorine zones between the Liao river and the Nenjiang river. The border in the north part of low fluorine zone was in Taonan county. In the area between the Zhenlai county and the Daan county, and the stretch of valley of Nenjiang river, there was 0.39 2.5 mg/l of fluorine concentration in unconfined ground water. The border of the south part of low fluorine zone was in the Tonyu-Changling-Qianguo counties, where the fluorine concentration was 0.21–1.5 mg/l. In some local areas, it was higher than 2.0 mg/l in special site only. High fluorine zones were mainly distributed in three large areas: one high fluorine zone of the southern part was from the west Zhanyu to the east Changling, Nong'an counties. here was 1.73–18.5 mg/l fluorine concentration in unconfined ground water. High fluorine zones of the northern part was distributed in the Baicheng area between the Taoer river and the Nenjiang river, and then towards to the east of Zhaoyuan, Zhaozhou, Zhaodong areas. It is 1.68–5.9 mg/l of fluorine concentration in unconfined ground water of the northern part area. Another high fluorine zone was in the middle area included the south part of Taoer river, the north part of Tongyu, the south part of Da'an-Qian'an and Qianguo county, where the fluorine concentration was 1.6–12.0 mg/l. From 3-D diagram (Fig. 2) found that the fluorine concentrations of unconfined ground water was gradually lower from the east or the southeast to the west or the northwest part in Songnen Plain, which was consistent with the contour map of fluorine concentration in unconfined ground water.

There are two aquifers in confined ground water, which is consisted of confined ground water in loose porous rocks of the Baitushan group of lower pliostocene, Quaternary and the Taikang group of upper Pliocene of the Tertiary (Liao and Lin 2004). Table 3 showed that fluorine concentrations in confined ground water were lower than that in unconfined ground water. This is why the aquifers of confined ground water are not affected by natural environmental factors in the earth surface. The hydrochemistry characteristics of the unconfined ground water were affected and limited by environmental and hydro-geological conditions from the recharge area to the sink area. The distribution character of fluorine concentration in the aquatic-environment was that the fluorine concentration in the lakes was higher than that in the river waters, and fluorine in unconfined ground water was higher than that in confined ground waters.

**Table 2** Correlation matrix of the fluorine with other chemical composition in main lake water

| | $F^-$ | $K^+$ | $Na^+$ | $Ca^{2+}$ | $Mg^{2+}$ | $Cl^-$ | $SO_4^{2-}$ | $HCO_3^-$ |
|---|---|---|---|---|---|---|---|---|
| $K^+$ | −0.491 | | | | | | | |
| $Na^+$ | 0.425 | 0.580 | | | | | | |
| $Ca^{2+}$ | −0.849 | 0.877 | 0.117 | | | | | |
| $Mg^{2+}$ | −0.881 | 0.845 | 0.054 | 0.998 | | | | |
| $Cl^-$ | 0.561 | 0.446 | 0.988 | −0.039 | −0.102 | | | |
| $SO_4^{2-}$ | 0.973 | −0.274 | 0.624 | −0.703 | −0.746 | 0.738 | | |
| $HCO_3^-$ | 0.638 | 0.358 | 0.968 | −0.135 | −0.197 | 0.995 | 0.80 | |
| $CO_3^{2-}$ | 0.870 | 0.003 | 0.816 | −0.478 | −0.533 | 0.896 | 0.961 | 0.935 |

## Discussions

### Fluorine source

Fluorine in the aquatic environment of the west part of the Songnen plain is mainly from the mineral components in the Daxing'an mountains, the Xiaoxing'an mountains, the Changbai mountains and fluvial and alluvial deposits from the watershed of the Songhuajiang river and the Liao river, as well as sedimentary deposits of the Songnen plain. Rocks and minerals containing fluorine in this region have been exposed to the earth surface for long time. Since the mineral containing fluorine in the rocks of this region were weathered, leached, migrated and enriched (Table 4), it is easy to be dissolved and has been the main sources for high fluorine in the water. The Songnen plain is a larger subsided basin and accumulation plain with the deposits as thick as 140 m. The deposits are consisted of mineral constituents of loam and clay containing high fluorine. For example, fluorine concentration can be as high as 1–5% in the gypsum minerals (Zhang et al. 1996).

### Factors affecting fluorine distribution

#### *Differences in natural landscape*

Fluorine in unconfined ground water is from the minerals and rocks containing high fluorine. Fluorine in the minerals and rocks in the highland is leached out and with the water flowing to the lowland and accumulated in the lowlands. Fluorine running directed to that unconfined ground water is from leached areas to migrated areas, and then accumulated areas due to the effect of the differences in natural landscape on the fluorine movement. It is consistent with chemical type of the ground water, i.e. from $HCO_3^-$–$Ca^{2+}$.$Mg^{2+}$,or $HCO_3^-$.$SO_4^{2-}$–$Ca^{2+}$.$Mg^{2+}$to $HCO_3^-$.$SO_4^{2-}$–$Ca^{2+}$. $Na^+$, or $HCO_3^-$.$CL^-Na^+$. $Ca^{2+}$and then to $HCO_3^-Na^+$, or $SO_4^{2-}Na^+$. The flow direction of the groundwater also is from recharge areas to runoff

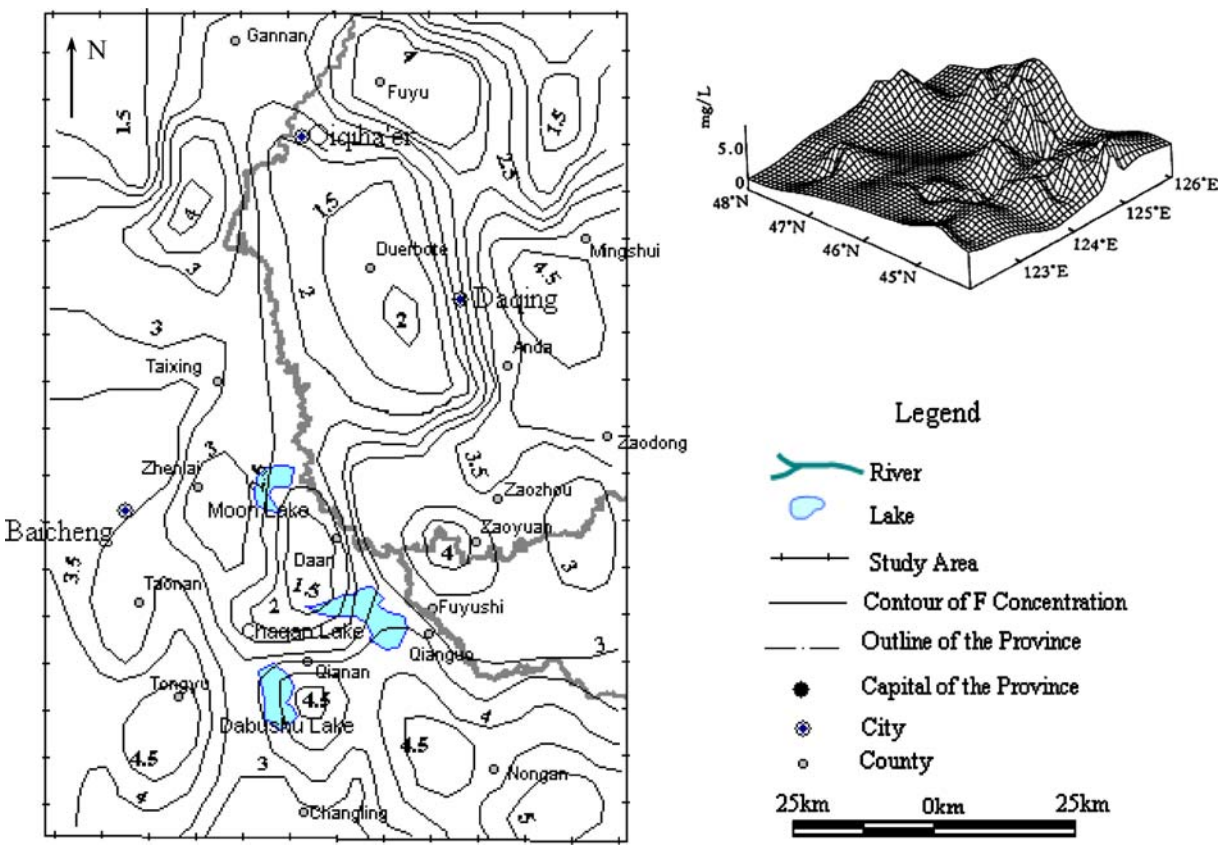


**Fig. 2** Sampling sites and fluorine distribution in unconfined groundwater, study region

**Table 3** Fluorine contents and other chemical characteristics in unconfined ground water and confined ground water (mg/l)

| Location | Water type | $F^-$ | $Na^+$ | $Ca^{2+}$ | $Mg^{2+}$ | $Cl^-$ | $SO_4^{2-}$ | $HCO_3^-$ | pH | TDS | Type of water chemistry |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Da an Haituo | Unconfined | 1.20 | 305.67 | 36.07 | 32.87 | 97.13 | 4.80 | 908.59 | 7.65 | 0.932 | $HCO_3^-$–$Na^+$ |
| | Confined | 0.98 | 121.90 | 47.09 | 25.24 | 64.87 | 13.93 | 462.53 | 7.50 | 0.529 | $HCO_3^-$–$Na^+$ |
| Da an Pin gan | Unconfined | 2.09 | 460.23 | 16.03 | 34.05 | 406.61 | 0.00 | 727.97 | 7.85 | 1.287 | $HCO_3^-$,$Cl^-$–$Na^+$ |
| | Confined | 1.08 | 182.16 | 60.12 | 36.48 | 146.05 | 70.60 | 504.03 | 7.50 | 0.749 | $HCO_3^-$,$Cl^-$–$Na^+$ |
| Daan Longzhao | Unconfined | 1.30 | 404.80 | 116.23 | 114.30 | 788.05 | 0.00 | 634.61 | 7.34 | 1.689 | $Cl^-$, $HCO_3^-$–$Na^+$$Mg^{2+}$ |
| | Confined | 0.30 | 129.72 | 17.64 | 0.00 | 46.79 | 10.57 | 335.00 | 7.50 | 0.415 | $HCO_3^-$–$Na^+$ |
| Qianan Dashi | Unconfined | 1.84 | 159.16 | 118.24 | 92.42 | 198.17 | 65.80 | 653.52 | 7.60 | 1.126 | $HCO_3^-$,$Cl^-$–$Mg^{2+}$,$Na^+$,$Ca^{2+}$ |
| | Confined | 0.80 | 167.44 | 66.53 | 26.51 | 98.04 | 80.21 | 518.06 | 7.60 | 0.693 | $HCO_3^-$–$Na^+$, $Ca^{2+}$ |
| Tongyu Shihuadao | Unconfined | 2.50 | 905.05 | 32.06 | 70.53 | 384.63 | 0.00 | 2180.85 | 7.77 | 2.558 | $HCO_3^-$–$Na^+$ |
| | Confined | 0.80 | 120.52 | 18.04 | 12.16 | 24.46 | 14.41 | 372.83 | 7.80 | 0.377 | $HCO_3^-$–$Na^+$ |

areas and then to sink areas. At the same time, fluorine concentration in unconfined ground water was changed from fluorine deficiency in the highlands (mountains and hills) to fluorine optimum between the highlands and lowlands, and then to fluorine excess in the lowlands.

*Climate*

It is clearly that there is a transitional climate in this region, where is from semi-humid to semi-arid and arid areas in the continent climate with the monsoon in the temperate zone (Liu 2001). Its precipitation is 350 mm in the west and the northwest parts and 500 mm in the east and the southeast parts. Annual evaporation is more than 1,500 mm and gradually decreased from the northwest to the southeast part. Evaporation is the main discharge path of the groundwater in the basin. Because the evaporation is much more than precipitation, especially in the spring, fluorine in unconfined ground water is much easily concentrated and resulted in high fluorine in unconfined ground water.

*Hydrochemistry characteristics*

The geochemical behavior of fluorine is controlled and limited by calcium in the environments (Hitchon 1995; Whiteford 1991). In order to discuss fluorine levels controlled by the calcium in unconfined ground water, regression equation analysis has been conducted in chemical components of unconfined ground water. It was assumed that $F^-$ was independent variable, while $Ca^{2+}$ and $HCO_3^-$ were variables in the unconfined ground water. It was clear that a log-correlation existed between fluorine and calcium concentrations in the regression equation: $[F]=7.62-1.48\ \ln[Ca]$, $r=0.988$, $p<0.001$. It was significantly negative correlation between fluorine and calcium. In addition, pH was a key factor because $Ca^{2+}$ and $HCO_3^-$ concentrations in unconfined ground water were controlled by pH, which results in fluorine enriching in alkali conditions.

**Table 4** Fluorine contents in several minerals and rocks (mg/kg)

| | Phosphorite | Fluorite | Granite | Flake stone | Shale |
|---|---|---|---|---|---|
| Fluorine concentration | 12,219 | 68,675 | 1,312 | 5,535 | 841 |

*Soil soda-salinization*

Because of a strong evaporation in the west part of the Songnen plain, the water in soil and the groundwater were transported into topsoil through the capillary pores waters and become the material source of soil soda salinization. The pH value was increased in the soil. Most of $Ca^{2+}$ and $Mg^{2+}$ in the soil colloids were exchanged by $Na^+$, resulting in high alkalization to soils (Zheng 1983). Reactions of soil colloids and fluorine ions were shown as following:

$$X-\mathrm{Ca}+2\mathrm{Na}^+ = X-\mathrm{Na}_2+\mathrm{Ca}^{2+}$$

$$X-\mathrm{F}+\mathrm{OH}^-- = X-\mathrm{OH}+\mathrm{F}^- \quad (X\ \text{soil colloid})$$

When soil soda-saltilization was produced, it could move into right as mentioned above two reactions. On the one hand, pH value of soil solution rose. On the

other hand fluorine was absorbed on soil colloid and was exchanged by $OH^-$ to be free fluorine ion ($F^-$) in the soil solution. It led to fluorine enriching in the soil solution (Zheng 1983).

## Effects of fluorine on human health

It is easily to have endemic fluorosis in local residents of study region because the fluorine concentration in unconfined ground water is high (>1.0 mg/l, WHO 1991). Unconfined ground water with high fluorine is used as drinking water in local residents. The sources of drinking water, methods of water storage and utilization, as well as the contribution of the high fluorine in dinking water to dental and skeletal fluorosis were investigated and determined (Moturi et al. 2002). For example, there were 1.8 million people in the Baicheng region and the Songyuan region of the Jilin province, the patients of dental and skeletal fluorosis were about 0.72 million people, and the morbidity of dental fluorosis and skeletal fluorosis in two regions was 39.47%. So the fluorine concentration in the groundwater was closely associated with human health, especially to children's health. Typical fluorosis areas, such as the Zenlai, the Taonan and the Fuyu counties were chosen to study the relationship between the fluorine concentration in drinking water and endemic fluorosis. There were positive correlations for the relationship between fluorine concentration in drinking water and the morbidity of dental fluorosis in the Zenlai county, the Taonan county and the Fuyu county. Correlation coefficient is $r_1=0.946$ ($p<0.01$), $r_2=0.518$ ($p<0.05$), $r_3=0.762$ ($p<0.01$), respectively. Among them, the correlation of fluorine concentrations in drinking water and the morbidity of dental fluorosis in the Zhelain county was shown in Fig. 3. Correlation coefficient was 0.896, $p<0.01$, it was very remarkable for relationship between fluorine concentration in drinking water and morbidity of dental fluorosis in Zenlai county. Also, there was a positive correlation for the relationship of fluorine concentration in drinking water and the morbidity of the skeletal fluorosis in the Zhelain county (Fig. 3), $r=0.815(p<0.01)$ and regression equation $y=-7.18+5.40x$, where $y$ is the morbidity of skeletal fluorosis, and $x$ is fluorine concentrations in the drinking water. Therefore, the human health was affected by high fluorine in drinking water.

## Conclusions

As mentioned above, the study region is a typical subsided basin where is formed by the fluvial and alluvial accumulations from the rivers and streams in the plain/ the mountains and hills around. Fluorine concentration in the river and stream water was lower than that in the lakes and shallow water. Fluorine concentration in confined groundwater was lower than that of unconfined groundwater in the study site. In unconfined ground water, from the east and southeast areas to the west and the northwest areas of the plain, fluorine concentration fluctuated with high and low alternatively. Fluorine source in aquatic environments is from weathering and dissolving of the rocks and minerals contained high fluorine in the plain/mountains and hills around. Main influence

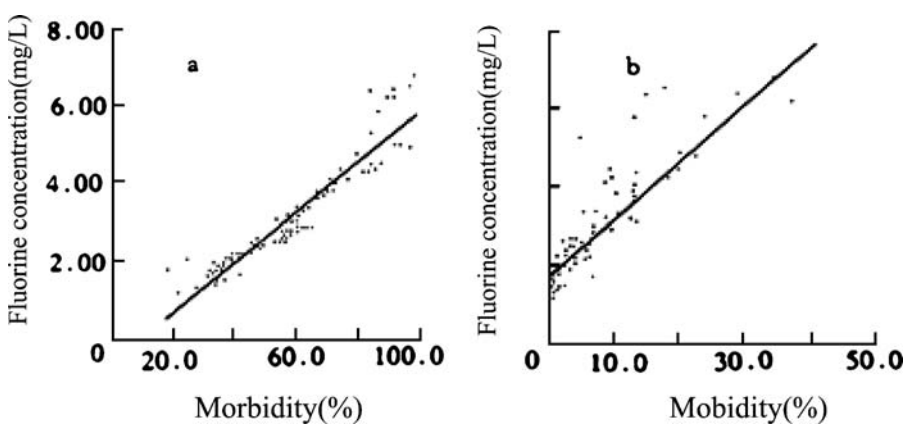


**Fig. 3** Correlation of fluorine concentration in drinking water and morbidity of dental fluorosis and skeletal fluorosis in Zenlai county (**a** dental fluorosis **b** skeletal fluorosis)

factors on fluorine distribution in aquatic environments were natural landscape difference, local climate, hydro-geological characteristics and strong soda salinization of soils. Unconfined groundwater containing high fluorine is main source of drinking water for local residents. Endemic fluorosis is apparent in local residents and threatened to the resident health, especially children health.

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