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Environmental Characteristics of Regional Groundwater in Relation to Fluoride Poisoning in North China

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ABSTRACT / More and more data indicated that high- or low-fluoride-bearing drinking water led to endemic diseases in which fluoride poisoning was caused by high levels of fluoride (fluoride ion content >1.0 mg/l) in drinking water. Fluoride poisoning in North China is characterized by pathological changes of bones and teeth. Much attention has been devoted to the study of fluoride-bearing groundwater in North China because regionally groundwater has been the main source of water supply, and fluoride poisoning has developed to the extent that it affects human health seriously. Results from the studies in North China summarized in this article indicate that regional high-fluoride-bearing groundwater has a regular distribution corresponding with the development of endemic fluoride poisoning and has something to do with paleogeographic and paleoclimatic conditions, geology, and hydrogeology, especially with types of hydrogeochemistry, pH value of groundwater, degree of mineralization, and so forth. High-fluoride-bearing groundwater in relation to fluorosis occurs mainly in North China, and many effective measures have been taken to reduce the fluoride content in drinking water and to cure the disease after analyzing the distribution and environmental characteristics of high-fluoride-bearing groundwater.

Introduction

Groundwater in North China has been a main source of water supply due to a shortage of surface water, although the groundwater resource in this area accounts for nearly 20 percent of the whole amount of the nation. Endemic disease characterized by pathological changes of bones and teeth had been found in several provinces of North China from the end of 1950s to the beginning of 1960s and was diagnosed as endemic fluoride poisoning (including fluor bone symptoms and endemic dental fluorosis) caused by high levels of fluoride in the drinking water. According to official statistical data, about 45 million persons, accounting for one-twentieth of the total population in China, drank high-fluoride-bearing water (Xu 1988). In recent decades endemic fluorosis has been found in large numbers of people in more than ten provinces of North China as programs have developed for prevention and cure of endemic diseases. For example, 5.7 million persons in 124 counties (cities) in Hebei Province are at risk of fluor disease and 1.6 million persons are suffering from the disease (Xu 1988). In Xinjiang Autonomous Region, those suffering from endemic dental disease account for 42.8 percent of a diagnosed 37 thousand persons; the patients with fluor-bone symptoms in areas of high disease occurrence account for 20 percent of those with fluoride poisoning (Cheng 1988). Some progress has been made in preventing and curing endemic fluorosis, for example, drugs, method of isolation, artificial treatment of drinking and irrigation water, using high-quality water sources, and so forth (Cheng 1988; Zhou 1989; Liu and Zhu 1989). Ren and Jiao (1988) have discussed the distribution and formation of high-fluoride groundwater in China comprehensively. The approach on endemic fluoride poisoning has been well documented from the angle of environmental science by comprehensive study by hydrogeologists and medical specialists. These research reports, however, have seldom described the relation between endemic fluoride poisoning and environment (especially geological environment) from the angle of the whole region of North China. Environmental characteristics of regional groundwater with high-content fluoride are summarized and discussed in this article according to the distribution of high-fluoride-bearing groundwater and the symptoms of endemic fluoride poisoning after a two-year field investigation and data analysis.

Paleogeography and Paleoclimate

High-fluoride-bearing groundwater is distributed mostly over arid and semiarid areas of North China. Under the conditions of the arid climate, the action of evaporation and concentration is strengthened, thus

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mutual action and exchange absorption action among ions are also strengthened. In the conditions of the semiarid climate movement of groundwater is slow and water exchange is low, and the distribution areas of weathering crust of granite and alkaline magmatic rock and area of soda salinized soil are advantageous to the concentration of fluoride ion (F^-) in groundwater.

The F^- concentration in groundwater is subject to control by latitude. The region of 35°–50°N latitude in China is divided as an evaporation–concentration zone and belongs to arid and semiarid areas with little precipitation (only 200–400 mm/yr) and a high evaporation rate. Soda salinization of soil in the region and F^- concentration in groundwater are due to high evaporation. For instance, the content of F^- of phreatic water in the Alashan Desert area reaches to 15.5 mg/l (Yang 1987).

The distribution of high-fluoride-bearing groundwater has different geomorphologic types. The geomorphology in Hebei Province is divided into four types: plain, basin, mountain, and highland areas (Liu and Zhu 1989). In plain area the soil is rich with carbonate sediment (CO_3) and Na(I); the content of aqueous fluoride increases, and fluoride-bearing materials that permeate the soils are leached by the infiltration of precipitation and are concentrated in the groundwater. In the highland area the content of F^- in groundwater indicates a decrease in concentration. Fluoride is concentrated in the groundwater of the center of Zhungeer Basin in Xinjiang Autonomous Region (Liu and Zhu 1989). In the alluvial-flooded laked deposit plain and desert area of the middle and the lower reaches of Kuiteng River groundwater is mostly evaporated, and thus the content of fluoride in groundwater is as high as 28 mg/l. High-fluoride-bearing groundwater in Shanxi Province is distributed in the center of Datong, Yi County, and Yunchen Basin (Zhou 1988; Liu and Zhu 1989). The south Habao Plain is a continental arid plain, and the content of F^- in groundwater is very high, with the highest content of 12–18 mg/l (He 1987). The distribution of F^- in the confined aquifer of the Xi'an area appears in the northeast in a galley shape and appears to correlate with structural fractures. The F^- in groundwater occurs in various concentrations in the west Jilin Plain, Yuling Basin in Hebei Province, Eershan Mountain in Neimenggu Autonomous Region, and Huaibei in Anhui Province. These fluoride-bearing groundwater regions are related to the geological rock with different fluoride contents, which is the material foundation of the extensive concentration of F^-. A closed basin fracture and strong evaporation within the basin are conducive to the development of continental salinization and thus form a typical fluoride-concentrated environment. Different geographical types have different zones of hydrogeochemistry, and the fluoride content in groundwater also shows obvious zoning. A typical example can be seen in Table 1 (Shen 1986).

In the outlet areas of high thermal groundwater and thermal springwater, fluoride was concentrated in basic thermal water because fluoride-bearing silicate quickens weathering under the influence of temperature and F^- is leached from the rock.

High-fluoride-bearing groundwater areas are common in volcanic areas, and groundwater bears fluoride due to volcanic activities since the Cenozoic Era and the dissipation of fluoride in hot gases.

Data indicate that high-fluoride-bearing groundwater areas are also endemic fluor disease areas. As the human body absorbs fluorine from the environment and most fluoride is assimilated from drinking water, a high content of fluoride in drinking water is the main cause of endemic fluorosis (Table 2). The hygienic standard of public drinking water in China stipulates that the suitable concentration of fluoride in drinking water is 0.5–1.0 mg/l.

Geological Characteristics

Bedrock containing fluorine-concentrated minerals is extensively distributed in the area of high-fluoride-bearing groundwater. The fluorine element in fluorine-concentrated minerals dissolves gradually in groundwater and becomes one of the main trace elements of groundwater due to influence conditions such as temperature, pressure, and physicochemistry, and so forth in the process of long geological age. For instance, granite is extensively distributed in the high-fluoride-bearing groundwater area of the south Zhungeer Basin (Liu and Zhu 1989), and the lithology mainly is composed of red potassic granite, speckled hornblende granite, granodiorite, hornblende–biotite granite, etc. The main fluorine-bearing minerals of these granites include hornblende (F^- = 5–7 percent), muscovite ($F^- \leqq 2.1$ percent) and black mica (F^- = 5.2 percent), the most general auxiliary minerals are fluorapatite ($Ca_5(PO_4)_3F$) in which F^- accounts for 1.23 percent and topaz, tourmaline, etc. The Fupin and Wutai groups of the Archean Group distributes at the eastern foot of Taihangshan Mountain and are composed of gneiss and marble. These rocks contain a high content fluorine and provide a material base for the fluoride-bearing concentration in groundwater at the lower reaches due to their weathering, denudation, and transportation. The high-

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Table 1. Hydrogeochemistry zones of fluoride in groundwater of evaporation–concentration type

Hydrogeochemistry zones	Area	Fluoride content (mg/l)		Hydrochemistry type
		Average	Range	
Leaching	Huinan	0.14	0.0–0.4	HCO_3–Ca
	Jilin	0.18	0.0–0.8	HCO_3–Ca
	Changchun	0.46	0.01–2.0	HCO_3–Ca
	Fuyu	0.44	0.01–2.0	HCO_3–Ca
Transition	Changlin	1.64	0.3–5.0	HCO_3–CaNa
Salinized	Ganan	10.18	0.8–30	HCO_3–Na

Table 2. Relation between endemic fluorosis and fluoride content in drinking water

Content (mg/l)	Investigated persons (*n*)	Dental fluorosis		Fluor bone symptoms	
		n	%	*n*	%
1.0–1.5	3,129	833	26.6	56	1.8
1.6–3.9	33,070	15,529	46.9	3,489	10.6
4.0–7.9	64,623	42,737	66.1	13,340	20.6
8.0–20.0	17,579	12,938	73.6	5,699	32.4
Total	118,401	72,037		22,584	

Table 3. Relation between deposited thickness of volcanic rock and fluoride content in groundwater

Water-bearing bed	Total deposited thickness of volcanic rock (m)	Content of fluoride in groundwater (mg/l)
I		0.1–2.0
II	54.20	4.8–6.0
	149.61	
III	158.14	3.5–6.0
IV		1.0–4.0

fluoride-bearing groundwater area in Huaibei, Anhui Province, is composed mainly of coal and carbonate rock strata.

Deposited materials of volcanic activities and volcanic rock, and fluorine-bearing rocks or minerals become one of principal sources of fluoride in groundwater. In Cangzhou, Hebei Province, the content of fluoride in peridotite accounts for 0.01–0.27 percent and in basalt 0–0.5 percent. The higher the deposited thickness of volcanic rock, the higher the content of fluoride in groundwater (Table 3).

Hydrogeological Characteristics

Hydrogeological setting and hydrogeochemical characteristics are the most important factors for the concentration of high-fluoride-bearing groundwater. The zonality of high-fluoride-bearing groundwater, the influence of hydrochemical types on the distribution of fluoride content, the relation of fluoride content of groundwater, degree of mineralization, and pH of groundwater are closely interrelated with the regional hydrogeological setting and hydrogeochemical characteristics.

Zonality of Fluoride-Bearing Groundwater

High-fluoride-bearing groundwater has the regularity of zonality, especially for the distribution of fluoride in phreatic water. The content of fluoride in phreatic water in a leaching zone of the mountain area is 0–0.4 mg/l; in the plain area 0.4–1.5 mg/l. The fluoride content of phreatic water in the continental salinized zone, however, is 0.8–3.0 mg/l and the maximum concentration is as much as several tens of milligrams per liter.

In many fluorine source areas, the distribution of fluoride content has horizontal zonality from the mountain area to the plain area. Groundwater in mountain areas has a high hydraulic gradient with good groundwater movement and high water exchange. The hydrochemical type of groundwater is mostly bicarbonate–calcium (HCO_3–Ca) with a small degree of mineralization and the content of fluoride in the groundwater is very low. With the increase of movement of groundwater the thickness and lithology of the aquifer changes, and the permeability decreases, and water exchange becomes slow. Meanwhile the exchange function of positive ions strengthens, sodium Na(I) in groundwater increases and replaces calcium Ca(II), and the content of fluoride in ground-

Table 4. Fluoride content (mg/l) of groundwater at lower reaches of Kuiteng River, Zhungeer Basin, Xinjiang

Site	Surface water	Shallow confined water	Deep confined water
126	<1.5	2.25	6.28
127	<1.5	5.11	10.30
128	<1.5	2.95	6.42

water increases. In the center of the basin, F^- is largely concentrated in groundwater due to the closed basin fracture and topographic relief, low permeability, slow groundwater movement, and enhancement of evaporation and concentration. From the mountain area to the plain, therefore, the content of F^- tends to gradually increase. For example, at the southern foot of Zhungeer Basin and in some large basins of Shanxi Province, high-fluoride-bearing groundwater typifies horizontal zonality (Wang and Li 1987).

The endemic fluoride diseases are closely interrelated with the characteristics of environmental hydrogeochemistry (Xiong and others 1987). In the mountain area with good groundwater movement in the east of Jilin Province, there were only a few mild cases of endemic dental fluorosis. In the western plain, with poor groundwater movement, however, those with endemic dental fluorosis reached 50–80 percent, and those with fluor bone symptoms reached over 1–5 percent of the population. In the acid environments the diseases are milder and in basic environments the diseases are more severe.

The vertical zonality of fluoride in groundwater has the following three conditions:

1. F^- content shows a tendency to gradually increase from top to bottom, which turns out to be contrary to the vertical zonality of common elements inversion. This circumstance appears in confined aquifers of high-fluorine-bearing areas and the content of fluoride in deep aquifers is higher than that in shallow aquifers (Table 4). This situation may be interrelated with the decrease and even disappearance of calcium Ca(II) content in groundwater, with the increase of sodium Na(I) content and with enlargement of F^- activity (rF/rCa). There is a similar condition in every large basin of Shanxi Province (Wang and Li 1987; Zhou 1989).

2. Different depths have different concentrations of F^- in groundwater. For example, the content of fluoride in the eastern plain of Hebei Province changes from small → larger → largest → smaller from the first aquifer to the fourth one; the largest content of fluoride appears in the third aquifer (Fig. 1).

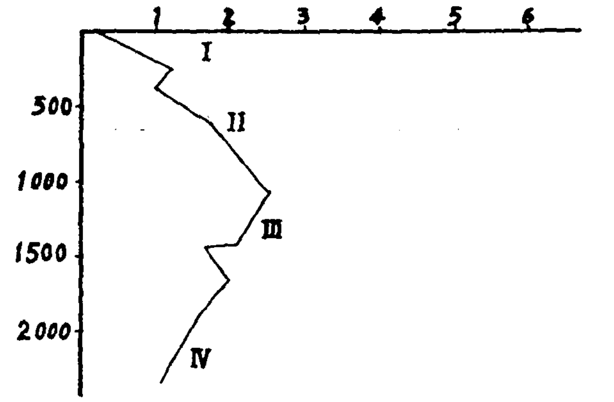

Figure 1. The vertical change curve of fluoride ion, Cangzhou, Hebei.

3. With the increase in depth to groundwater, the content of fluoride decreases due to different stratum age and different types of loose layer. For example, the high-fluoride-bearing groundwater area in Yuncheng Basin, Shanxi Province, was a salty concentration zone in the fracture basin of the Cenozoic Era. A thick unconsolidated alluvium and lake sediments were accumulated within the basin. The middle to bottom layers are composed of clay, clayey soil, sand soil, powdery soil, and fine sand; the upper layers are loess accumulation (Wang and Li 1987). According to the geological structure explanation of satellite photography of South Shanxi there is a concealed fracture oriented NNE–SSW in the center of the basin. The content of fluoride in the concentration zone increases because of the dissolution and infiltration of groundwater in mountain area, inclined flood plain, and alluvial plain. At the upper layer, the particulates are fine, phreatic water is shallow, and evaporation capacity is large, which cause phreatic water concentration and the content of fluoride ion to rise to form the present high-fluoride-bearing groundwater area (Table 5). According to drilling data, the stratum between 0 and 400 m can be divided into the following water-bearing groups. Q_{2+3} water-bearing group is buried under 80–100 m and the thickness of the water-bearing layer is 15–25 m. The edge of the basin is composed mainly of the medium–fine sand. Toward the center, which is composed mainly of powdery and fine sand, the particulates are thinner than along the edge. The degree of mineralization is 1–2 g/l, permeability coefficient is 4.8–10.2 m/day, water discharge is 60–84 m^3/day, and the F^- content is 4–10 mg/l. This group was once the principal source of drinking water, and the symptoms

Table 5. Relation between fluoride ion content and groundwater depth

Sites	Content (mg/l)[a]	Well depth (m)	Content (mg/l)[a]	Well depth (m)
Qiji	5.4–6.2	20–40	1.0	75–120
Miaoshangteng	2.5	15	1.4	110–147
Fengjiazhuo	9.0	30	3.4–6.0	100
Xirenshang	3.0	35	0.75	150
Renli	5.1–7.2	16	0.8	190
Xixia	6.0	20	3.25	120
Xikaizhang	1.9	40	0.77–0.88	110
Xibei	3.25–5.30	15–20	1.80	110–200
Xialuzhi	2.65	24	0.42	120
Houjiazhuo	7.4–8.0	15–18	1.12–1.36	107–135
Shangyi	3.1–7.0	3.6	1.54	300
Dajin	3.4–3.5	5–6	1.16–1.3	160–190
Sibei	5.0	30	1.04	260

[a]The fluoride ion content was measured with the fluoride electrode method.

of fluor bone disease are serious. The upper water-bearing group of Q_1^2 is buried under 80–120 m, 120–180 m in the south and 160 m in the west. The thickness of the water-bearing layer is 40–70 m; the lithology is mainly unconsolidated medium–thick sand. This group is one of the principal water-bearing layers in the basin and is also the principal source of drinking water. Degree of mineralization is 1–2 g/l, water discharge is 86–172 m^3/day, permeability coefficient is 0.2–0.5 m/day, and the concentration of fluoride ion is 0.8–1.5 mg/l. The bottom water-bearing group of Q_1^2 is buried under 160–240 m; the lithology is mainly powdery and fine sand, permeability coefficient is 20 m/day, water discharge is 480–720 m^3/day, degree of mineralization is 1–1.50 g/l, and the concentration of F^- is 0.7–1.0 mg/l. It is thus clear that fluoride ion concentration is different and related to different stratum age and different types of unconsolidated layers. In general, the content of F^- in $Q_{3+2} > Q_1$, clay and clayey soil > sand soil > fine sand.

Relation Between Hydrochemical Types and Fluoride Content

As hydrochemical types are mainly hydrocarbonate–calcium (HCO_3–Ca) water and hydrocarbonate–calcium sodium (HCO_3–CaNa) water, the concentration of F^- in groundwater is low. As hydrochemical types gradually change to hydrocarbonate sulfate–sodium calcium (HCO_3SO_4–NaCa) or hydrocarbonate–sodium (HCO_3–Na) and sulfate–sodium (SO_4–Na) water, the concentration rises. Groundwater of the hydrocarbonate–sodium (HCO_3–Na) type always has a very high fluoride content, because the concentration of elements in groundwater depends on their solubility becoming certain chemical forms in groundwater and the solubility of sodium fluoride (NaF) is very high. In these types of groundwater, the fluoride exists in the form of a simple negative ion (F^-), which is the most active. Na(I) is one of the most active positive ions, and the combined power of F^- and Na(I) far exceeds their respectively mainly positive ions. Although the combination of F^- and Na(I) forms water-soluble salt, the content of fluoride ion increases relatively because the opportunity of sedimentation of calcium fluoride (CaF_2) is decreased greatly due to the containment of Na(I) in groundwater.

In the Yuncheng Basin of Shanxi Province, the content of F^- has a certain regularity with hydrochemical types in the respect of horizontal and vertical zonality (Wang and Li 1987). When groundwater quality in the zone of active water exchange is hydrocarbonate–calcium sodium (HCO_3–CaNa) or hydrocarbonate–calcium magnesium (HCO_3–CaMg), the content of F^- is <1.1 mg/l. The content of fluoride ion of hydrocarbonate sulfate (HCO_3SO_4) or sulfate hydrocarbonate (SO_4HCO_3) type water in the zone of decrease of groundwater movement condition is 1.1–1.5 mg/l. Fluoride ion content of hydrocarbonate chloride (HCO_3Cl) or sulfate chloride (SO_4Cl) and chloride-sulfate ($ClSO_4$) type water in the bottom of alluvial plain is 1.0–3.0 mg/l. In the confluence center of groundwater in the alluvial plain and low-lying land, the hydrochemical type is sulfate chloride (SO_4Cl), chloride sulfate ($ClSO_4$), and chloride (Cl). Fluoride ion content is 3.0–5.0 mg/l or >5.0 mg/l; the highest content reaches 15 mg/l in the zone of alluvial lake sediment.

Relation Between Fluoride Content in Groundwater and Degree of Mineralization

Data indicate that the fluoride content of groundwater increases with the increase in degree of mineral-

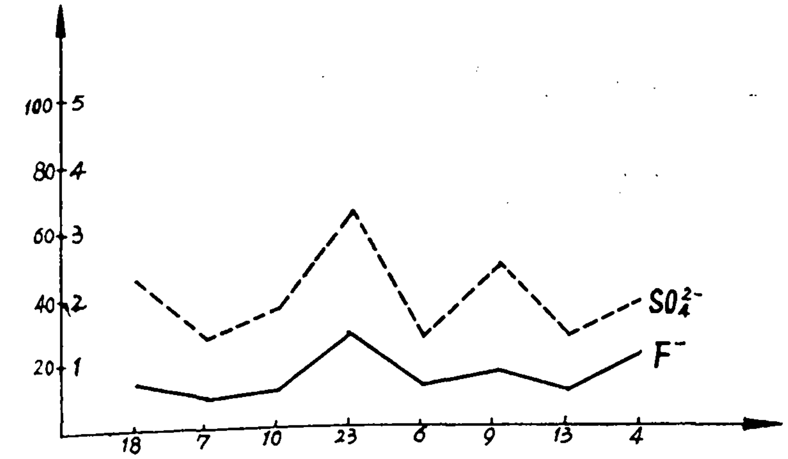

Figure 2. Correlation curve of sulfate (SO_4^{2-}) and fluoride ion content.

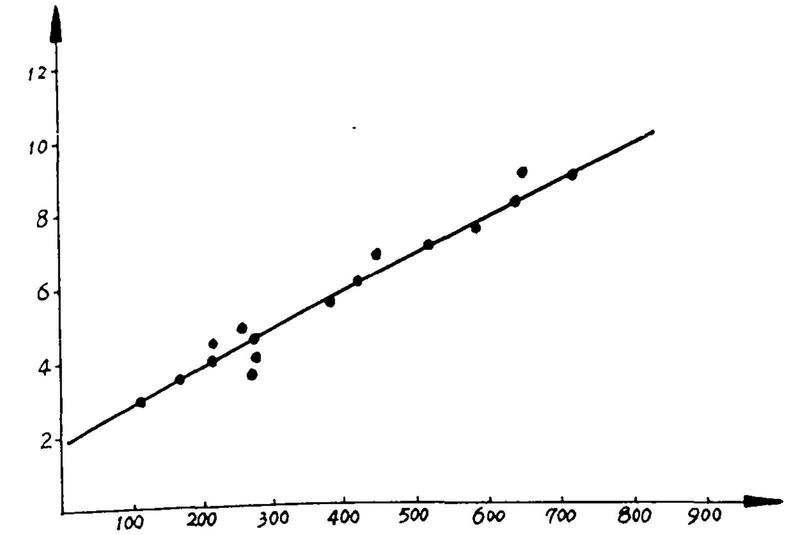

Figure 3. Correlation chart of F^- and Na(I).

ization. In the Zhungeer Basin as the degree of mineralization of phreatic water rises to 8.8–29.97 g/l from 0.08–0.2 g/l, the fluoride ion content increases to 2.69–5.37 mg/l from 0.11–0.2 mg/l. In the second water-bearing layer of the Quaternary System of the middle to the eastern Hebei Plain, Beichengzi area of Jilin Province, and every large basin of Shanxi Province, this relationship always holds, that is, the increase in sulfate content corresponds with fluoride content increases when the degree of mineralization increases. For instance, this kind of relation in Huaibei, Anhui Province, is very obvious (Fig. 2). When high fluoride-bearing groundwater mostly occurs with mixed type water and Na(I) is the predominant positive ion in this groundwater, the content of fluoride has a linear correlation with the content of Na(I) (Fig. 3).

The regularity of concentration of fluoride in Yuncheng Basin relates to the amount of Ca(II), K(I), and Na(I). This can be seen by the analytical data (Zhou 1989); fluoride content in Jian village is 4.0 mg/l, Ca(II) = 2.0 mg/l. The content of K(I) + Na(I), however, increases to 384 mg/l; the specific value of [K(I) + Na(I)]/Ca(II) is 192 mg/l. In the drilling well of Kangshu, fluoride content is 0.8 mg/l, Ca(II) content reaches to 34.1 mg/l, the content of K(I) + Na(I) is 88.6 mg/l, and the ratio of [K(I) + Na(I)]/Ca(II) is 2.59. The basic regularity is that the concentration of fluoride in groundwater increases with the increase in ratio of [K(I) + Na(I)]/Ca(II); it is inversely proportional to the content of Ca(II) and directly proportional to the content of K(I) + Na(I).

Exceptions exist because the fluoride content of groundwater is restricted by many factors. In the condition of the same degree of mineralization, the content of fluoride in groundwater is different. Sometimes when the content of fluoride is high, the degree of mineralization is not high. This phenomenon is obvious in high-temporature groundwater or mineral-

Table 6. Relation between fluoride content in phreatic water and pH value and hydrochemical types

Well	Fluoride content (mg/l)	Degree of mineralization (g/l)	pH	Hydrochemical type
1	1.00	0.99	7.6	HCO_3SO_4–CaNaMg
2	1.26	1.61	7.6	SO_4ClHCO_3–NaMgCa
3	3.31	1.70	7.7	SO_4HCO_3–CaNa
4	4.68	9.11	7.6	SO_4CL–NaMg
5	1.95	1.35	8.1	SO_4Cl–MgNaCa
6	2.82	1.96	8.1	$ClSO_4$–NaMg
7	1.12	4.88		
8	2.29	5.78	7.8	$ClSO_4$–Na

ized water. The degree of mineralization of most mineral springs in Guangdong Province is not high (0.2–0.4 g/l), but the content of fluoride ion can be as much as 10–20 g/l.

Relation Between Fluoride Ion Content and pH Value of Groundwater

The content of fluoride ion is related to the pH of the groundwater. Fluoride ion can be concentrated under the condition of acidity and basicity. When pH = 6.5–7.0, $F^- > 1.5–3$ mg/l. The larger the ratio F/Ca, the higher the fluoride content. Under this condition the environment of hydrogeochemistry is basic hydrocarbonate–sodium (HCO_3–Na) type water; the content of Ca(II) is low and Na(I) is very high.

In high-fluoride-bearing areas of Yeerwu River Basin, Xinjiang, the hydrogeochemical type of the aquifer of the Quaternary System is complex; hydrocarbonate–calcium (HCO_3–Ca) water changes to hydrocarbonate sulfate–calcium sodium (HCO_3SO_4–CaNa) water and sulfate chloride–sodium magnesium (SO_4Cl–NaMg) water, exchange of positive ions strengthens, Na(I) and Mg(II) in groundwater increase and replace Ca(II), and groundwater is in the condition of basicity (Table 6). Because fluoride transfers in the form of ion in the basic medium, the concentration of fluoride ion is directly proportional to pH value and the content of Na(I) and inversely proportional to the content of Ca(II).

Discussion and Conclusion

1. Fluoride in groundwater transfers and concentrates in specific geochemical environments under the comprehensive function of paleogeography, paleoclimate, geology, and hydrogeochemistry, etc.

2. High-fluoride-bearing media are the base of leaching and concentration of fluorine through groundwater and also the main reason for formation of high-fluoride-bearing groundwater. Groundwater cannot contain higher fluoride without this base.

3. Low groundwater movement is an important hydrodynamic condition in which fluoride concentrates in groundwater. Relevant geographic and geomorphologic positions are mostly overflow zone or subsidence zone of groundwater in the forward positions of alluvial fan and flood basin.

4. Arid and semiarid climate causes high evaporation of groundwater, which is an important factor in concentrating salts (including fluoride) in groundwater. Relevant strata related to the evaporation and concentration of groundwater are unconsolidated sediment with fine particulates and strong capillary action.

5. In general, basic hydrocarbonate–sodium (HCO_3–Na) water with higher pH value is advantageous to the concentration of fluoride.

6. Chemical form of fluoride existing in the environment is very important to the growth and decline of endemic fluor disease. For example, in the western plain of Jilin Province, fluoride poisoning is very serious when fluoride exists in the form of calcium salt. The chemical form of fluoride, therefore, should be determined; it may be the decisive link in revealing the relation between endemic fluoride poisoning and environmental hydrogeology.

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