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Geo-Environmental Factors' Influence on the Prevalence and Distribution of Dental Fluorosis: Evidence from Dali County, Northwest China

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Abstract: Residents living in areas with high fluorine environmental background will ingest excessive fluorine from the atmosphere, drinking water, food and other ways. Long-term effects of fluorine on the human body could cause people suffering from dental fluorosis and influence the sustainable development of the severe fluorosis areas. Previous studies have independently discussed the high fluoride environment in Dali County from the aspects of natural environment, drinking water quality and endemic fluorosis. This study carried out a detail investigation on dental fluorosis population in seven selected villages of Dali County, Shaanxi province, northwest China. The highest dental fluorosis index of 1.9 was found in Lianjia village located near the Anren depression, while the lowest dental fluorosis index of 0.0 was found in Jiaxi village near the Yellow River alluvium. Groundwater fluorine contents the range was 0.01 mg/L to 11.80 mg/L, with the highest value (2.6 mg/L) being observed in the 2nd terrace of Weihe River. The lowest groundwater fluorine content (0.8 mg/L) was observed in the Yellow River alluvium. Soil fluorine contents ranged from 1.18 mg/kg to 13.70 mg/kg, with its highest value (13.70 mg/kg) observed in Xinfeng village near the Anren depression. The lowest value of fluorine (1.18 mg/kg) was found in soil from the 1st terrace of Weihe River. As for the fluorine contents of corn, they ranged from 4.04 mg/kg to 7.72 mg/kg. The highest value (7.72 mg/kg) appeared in the 3rd terrace of Weihe River and the lowest value (4.04 mg/kg) in the 2nd terrace of Weihe River. The soil leaching was the dominant fluorine source of groundwater environment. Areas with severe dental fluorosis are located at the edge of the depression and the conjunction between steep slope and gentle slope. A poor correlation was found between the dental fluorosis index from the seven investigated villages and the corn fluorine content contrarily to the groundwater fluorine content, which positively correlated to the dental fluorosis index. Based on the obtained results, two recommendations were done to prevent and control dental fluorosis and accelerate the sustainable development in Dali County: to strengthen the use of low fluorine groundwater for drinking water supply, and to widely install the public water purifiers in the rural communities for purifying high-fluorine water to reduce the incidence of dental fluorosis in the population.

Keywords: dental fluorosis; environmental geology; groundwater; corn; soil

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Citation: Yang, M.; Zhao, A.; Ke, H.; Chen, H. Geo-Environmental Factors' Influence on the Prevalence and Distribution of Dental Fluorosis: Evidence from Dali County, Northwest China. *Sustainability* **2023**, *15*, 1871. https://doi.org/10.3390/su15031871

Academic Editor: Katarzyna Sygit

Received: 14 December 2022
Revised: 14 January 2023
Accepted: 16 January 2023
Published: 18 January 2023

1. Introduction

During the long-term life of human beings, the influence of geological environment on human health follows us throughout our lifetime [1]. Chemical elements in the geological environment are not only the material basis for the basic composition of human body, but also the source of nutrients for maintaining life activities. Indeed, human physiology is sensitive to the concentration of required elements and generally keeps in a stable range, not the more the better. Lack or excess of chemical elements in water and food may cause certain pathological changes in the human body [2–4]. Fluorine is one of the essential trace elements in the human body. It is mainly distributed in bones, teeth, nails and hair. Its effect on the body changes with its amount of intake. Dental fluorosis, a geochemical disease occurring in specific geographical and geological settings, is due to excessive intake and accumulation of fluoride by people living in a high fluoride environment through water, atmosphere or food [5]. Dental fluorosis is a very widespread geochemical disease, which involves more than fifty countries and regions worldwide, including China, India, Bangladesh, Vietnam, Sri Lanka, US, Canada, Egypt and South Africa. Five billion people live in high-fluoride environments, and the fluorosis population has reached 200 million all over the world. According to statistics, with the exception of Shanghai and Hainan, 29 other provinces, autonomous regions and municipalities in China have endemic regions of fluorosis, with a population of 116 million threatened, 40 million patients with dental fluorosis, and more than 2 million patients with skeletal fluorosis [6]. Fluorosis in arid and semi-arid areas of northern China is mainly caused by fluorine-rich weathering compounds or by runoff migration. The accumulation and concentration of fluorine-rich weathering compounds in flat and low topographic areas results in high fluorine background of the area. It indicates that fluorine diseases in arid and semi-arid areas originate from the enrichment of fluorine elements in the environment. The enrichment of fluorine is controlled by a variety of factors in the geological environment of specific areas (e.g., climate, hydrology, topography, hydrogeological conditions and geochemical environment) during long-term interaction [7]. Dali County is one of the areas where endemic fluorosis is more serious in Shaanxi Province. People in the area north of Luohe River suffer from dental fluorosis, bone fluorosis to varying degrees, and some even lose their ability to work [8]. Numerous researchers have independently studied the high fluorine environmental problems in Dali County from the perspective of the natural environment [9–12], the quality of drinking water [13–15] and endemic fluorosis respectively, indicating an interference of endemic fluorosis for regional sustainable development [16–18]. Because of the barriers between different specialties, the mechanisms and relationships between the geo-environmental factors and endemic fluorosis were ignored to comprehensively studied and discussed.

In this study, seven villages with different fluoride backgrounds were selected for the dental fluorosis survey, with the dental fluorosis index calculation. In parallel, groundwater, soil and corn were sampled systematically in view determining their fluorine contents. This enabled discussing in detail the relationship between the dental fluorosis index and groundwater and corn fluoride contents as well as the effects from soil, terrain and hydrogeological factors on the formation of groundwater with high fluoride contents. The aim of this study is to analyze the effect of fluorine levels in geographical and geological environment on the physical health of the population by investigating the fluoride contents in the groundwater, soil and corn as well as its dominant influence factors of the environment and in combination with the dental fluorosis index values in seven different villages. Finally, the countermeasures to alleviate the dental fluorosis of the population are proposed through geological and environmental characteristics to help sustainable development in the study area.

2. Materials and Methods

2.1. Study Area

Dali County is located in the eastern margin of Guanzhong Basin, Shaanxi Province, in the confluence area of the Yellow River, Luohe River and Weihe River. Its geographic coordinates are E 109°43′~110°19′, N 34°36′~35°02′, with a total area of 1766 km^2. The study area belongs to warm temperate semi-humid and semi-arid monsoon climate. The topography is high in the north and low in the South with elevations from 329 m to 534 m. The study area is mainly covered by Quaternary sedimentary strata with seven types of landforms. The landforms from north to south are sequentially the Loess Plateau (I), the Weihe 4th terrace (II), the Luohe river terrace (III), the Weihe 3rd terrace (IV), the Weihe

2nd terrace (V), the Weihe 1st terrace (VI) and the Yellow River alluvium in the Southeast (VII) (Figure 1).

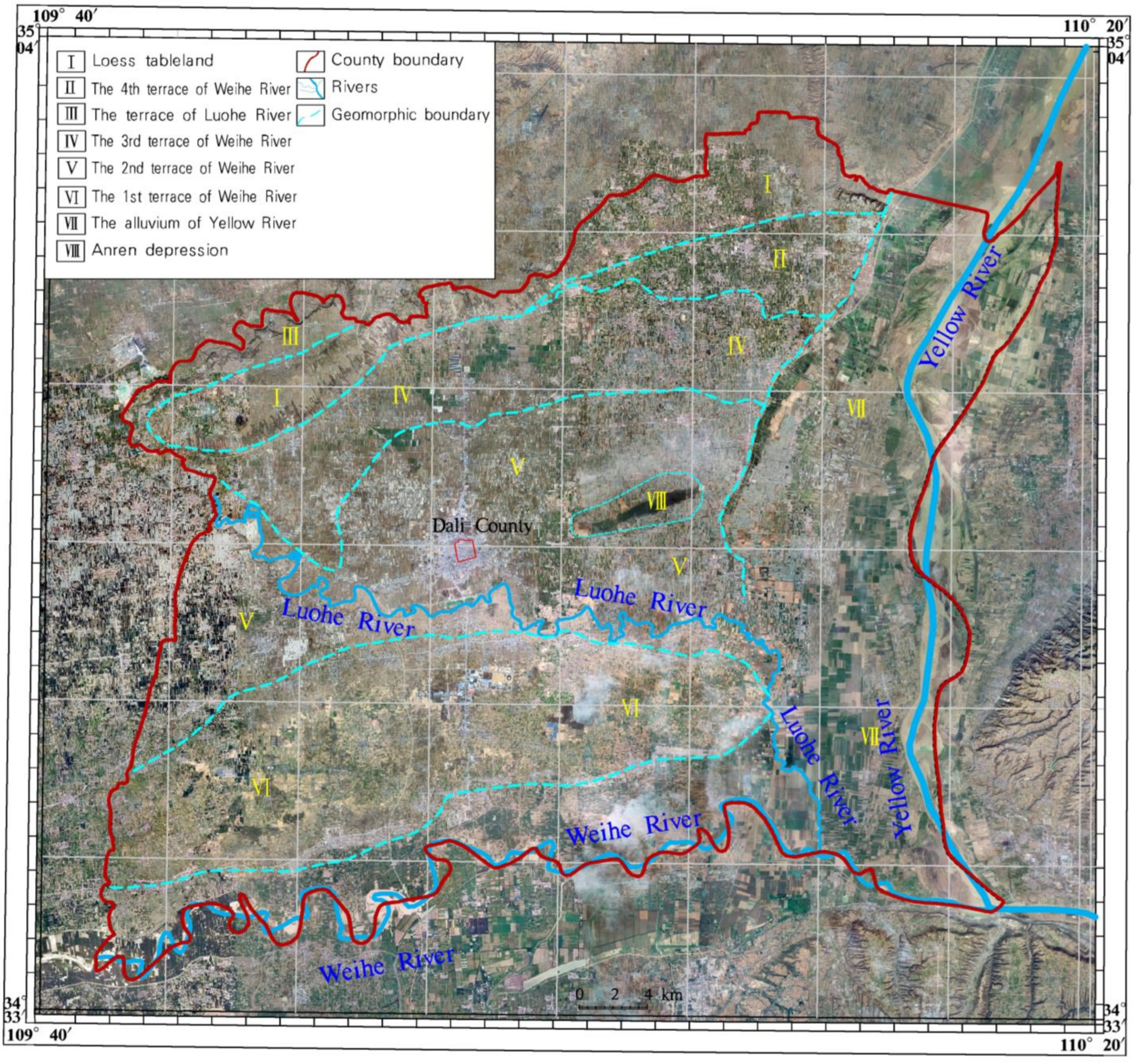

Figure 1. The geomorphic map of Dali County.

The research area is rich in water resources with an annual average precipitation of 514 mm. Groundwater flows roughly from north to south and west to east. In the western plateau area, groundwater flows from the middle of the plateau to valleys and terraces on both sides. The recharge of groundwater system mainly depends on atmospheric precipitation, confluence, irrigation infiltration and buried karst water [19]. Groundwater discharge mainly includes artificial exploitation and diving evaporation. The depth of groundwater in the whole area gradually becomes shallow from north to south. The maximum depth of groundwater about 110 m distributed in the northern plateau region and the groundwater depth over 50 m is located all over the study area [19]. The groundwater depths in the second and third terraces of Weihe River are from 2 m to 30 m [19]. The groundwater depths in the first terrace of Weihe River are from 8 m to 25 m in the south of the study area [19]. The groundwater depths of the Yellow River alluvium in the eastern part of the study area are from 3 m to 6 m [19]. Although the study area is rich in groundwater, nearly two-thirds of the groundwater are high-fluorine water and brackish water, which cannot be exploited for drinking. In addition, the contradiction between supply and demand of water resources in Dali County has become imbalance prominent due to the overgrowing water resources exploitation [20].

Dali County is a typical section of high-fluorine groundwater in Guanzhong Basin [10]. The rural population drinking high fluorine water is 187,800, accounting for 29.46% of the total rural population in Dali County [20].

2.2. Overview of Endemic Fluorosis in Study Area

According to the data of fluorosis census in 1980s and endemic fluorosis survey in 2006 in Dali County, over 276,600 fluorosis patients accounting for 57% of the total population of the disease area were tallied in the whole county [21]. Severe disease areas are distributed in the Anren depression whereas moderate disease areas are on the 2nd, 3rd and 4th terraces of the Weihe River. As for light disease areas, they are located on the Loess tableland and the south side of Luohe River. From the 1980s to 2015, the local water conservancies and Centers for Disease Control and Prevention have carried out effective fluorine prevention and water improvement work [16,22]. After the endemic diseases census of Dali County in the year 2018, only thousands of individuals with endemic fluorosis disease were newly sifted out, which is a significant reduction of the growth rate than the 1980s [13,23].

2.3. Methods

2.3.1. Investigation of Dental Fluorosis

The formation of dental fluorosis is determined by the amount of fluoride ingested during the development of permanent tooth crowns, generally around the age of birth to 5 years old [24]. The amount of fluoride ingested performs little effect on the teeth when the crown development is completed, that is, 5 years after birth [25]. Seven villages distributed in different geomorphic areas were selected for dental fluorosis survey for juveniles under 16 years old (Figure 2). A total of 529 juveniles were diagnosed with dental fluorosis using Dean's method [23]. The dental fluorosis index (DFI) was used to describe the prevalence of dental fluorosis in a given area. The index was calculated and used to achieve the classification according to China's diagnostic standard for dental fluorosis (WS/T 208-2011) [26]. Generally speaking, close relationships exist among the prevalence of dental fluorosis, water fluorine concentration and fluoride ingestion. The dental fluorosis index was calculated according to Formula (1), and the decision criteria were used as follows to make the classification: below 0.4 is negative; 0.4 to 0.6 is the normal range; 0.6 to 1.0 is mild prevalence; 1.0 to 2.0 is moderate prevalence; 2.0 to 3.0 is severe prevalence; above 3.0 is considered as very severe prevalence (WS/T 208-2011) [26].

$$\mathrm{DFI} = \frac{(\mathrm{N}_1 \times 0.5 + \mathrm{N}_2 \times 1.0 + \mathrm{N}_3 \times 2.0 + \mathrm{N}_4 \times 3.0 + \mathrm{N}_5 \times 4.0)}{\mathrm{N}_{\mathrm{Total}}} \quad (1)$$

where N_1 is the number of suspected cases; N_2 is the number of very mild cases; N_3 is the number of mild cases; N_4 is the number of moderate cases; N_5 is the number of severe cases.

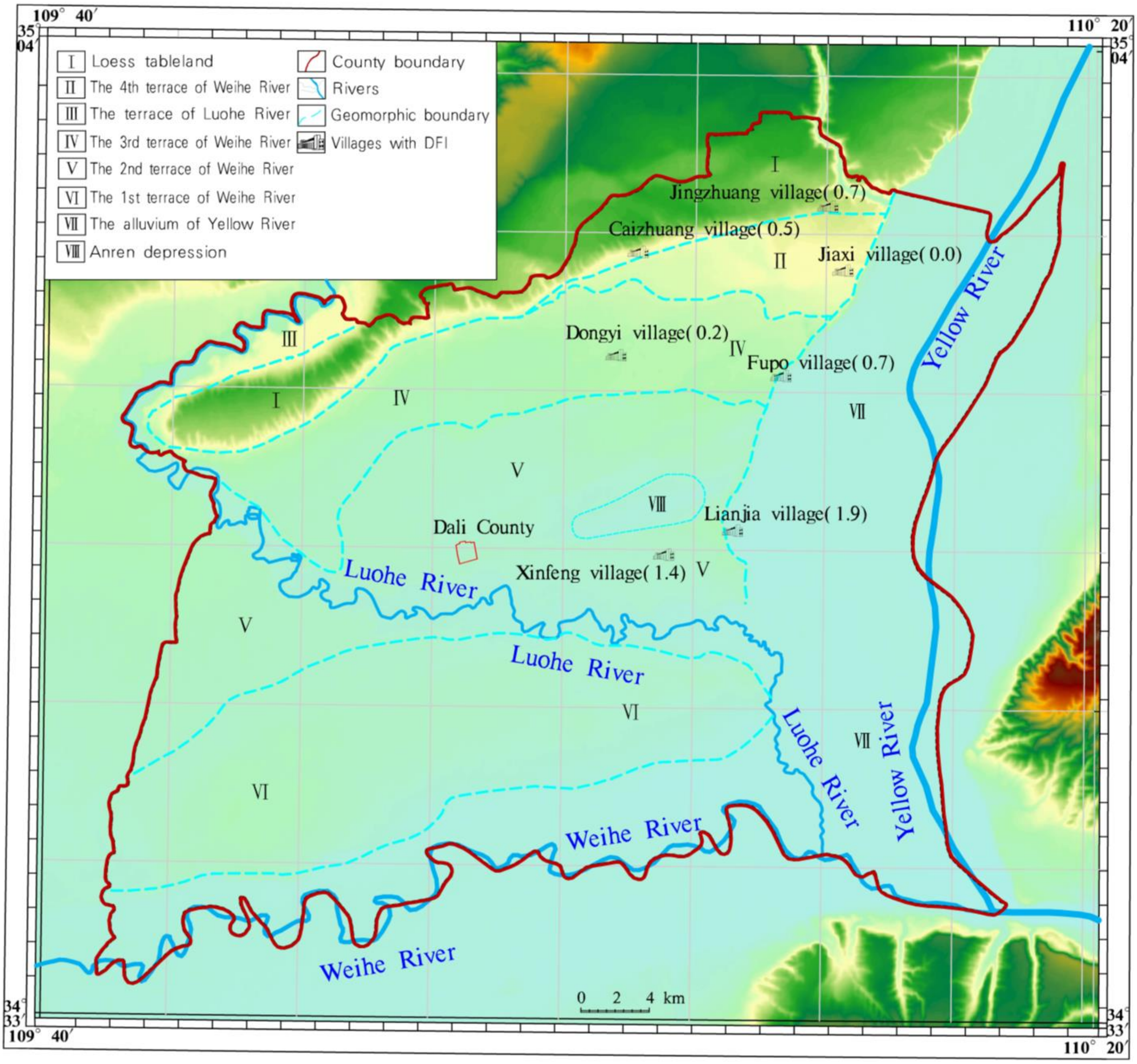

Figure 2. The distribution map of the survey villages for dental fluorosis.

2.3.2. Groundwater Sampling and Analysis

On the basis of comprehensive consideration of factors such as geology and landforms, groundwater exploitation and distribution of dental fluorosis in the study area, dense groundwater samples were emplaced from well water buried to within 90 m of depth as well as from a small area of spring water (Figure 3). The polyethylene plastic bottles with volumes of 2 L were used for sampling. The samples were sent to the laboratory within 48 h after collection. Fluorine anion contents were determined using an ion-selective electrode (ISE) according to the China National Standard for drinking water analysis (GB/T 5750.2-2006) [27]. This study excluded samples collected after rainfall and affected by pollutants such as pesticides and garbage. A total of 269 groundwater samples were selected for laboratory tests conducted at Xi'an Geological Survey Center of China Geological Survey, of which the laboratory has been awarded national metrology certification and national test and accreditation council accreditation approval.

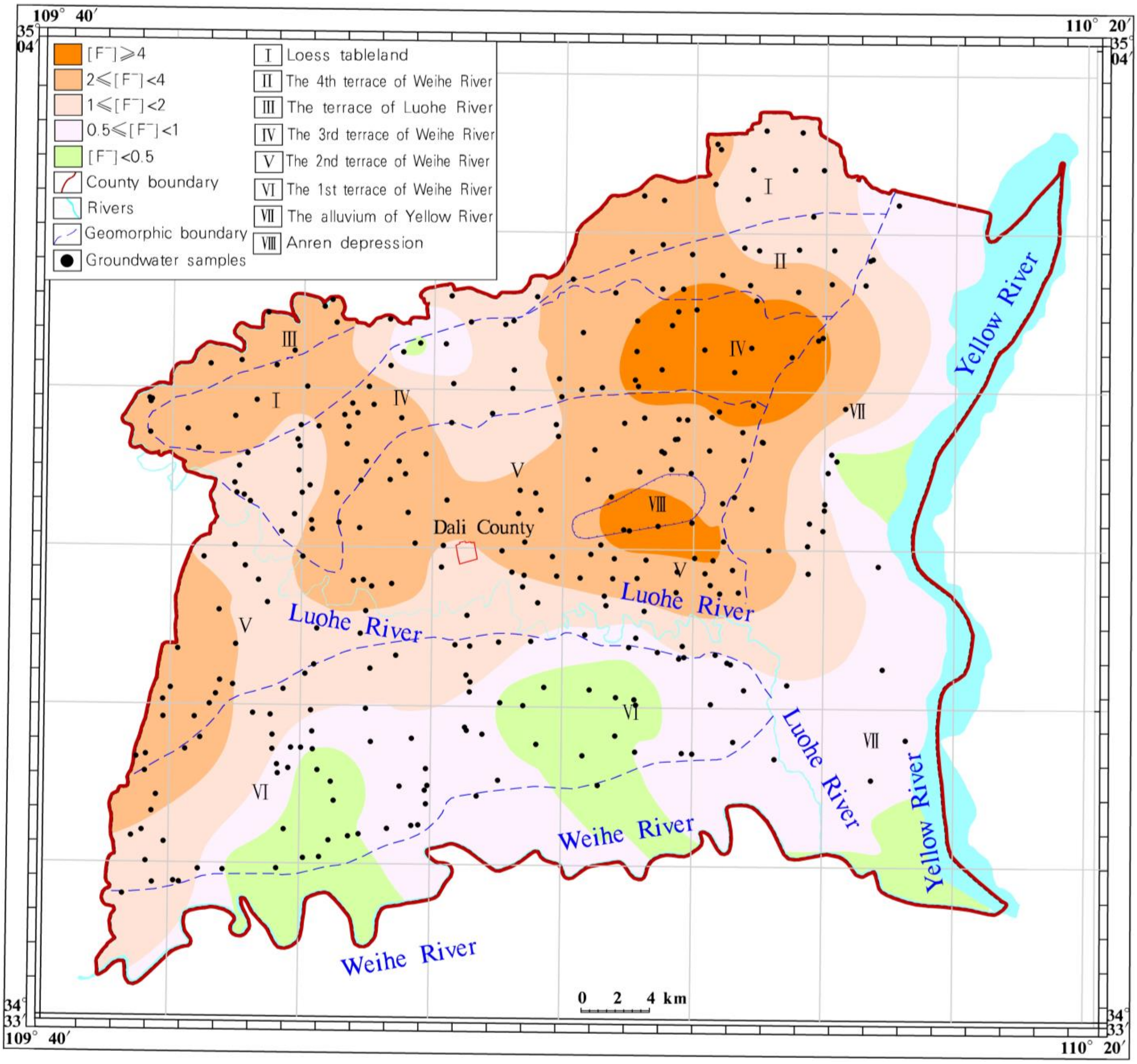

Figure 3. The distribution map of the groundwater fluorine contents.

2.3.3. Soil Sampling and Analysis

In this study, a total of 44 soil samples were collected in different geomorphic areas including Loess plateau, the Weihe terraces, the Yellow River alluvium, the Luohe terrace and the Anren depression (Figure 4). Five or six samples were collected from each geomorphic area, respectively. The area of each sampling site was 20 × 20 m, with three sample points selected in each plot using the diagonal cloth point method. The surface soil was collected from a depth of 20 cm and soil samples were stirred evenly using the tetrad method to take 1 kg into the sample bags. The samples were dried indoors under natural wind and ventilated from sunlight, and stone grains and plant residues were removed. After grinding and sieving (using a 100 mesh sieve), about 500 g were weighted in a sealed bag for further test. Soil samples were mainly tested for total fluoride content, with a relative error below 10% and the relative deviation of two independent results under 10%.

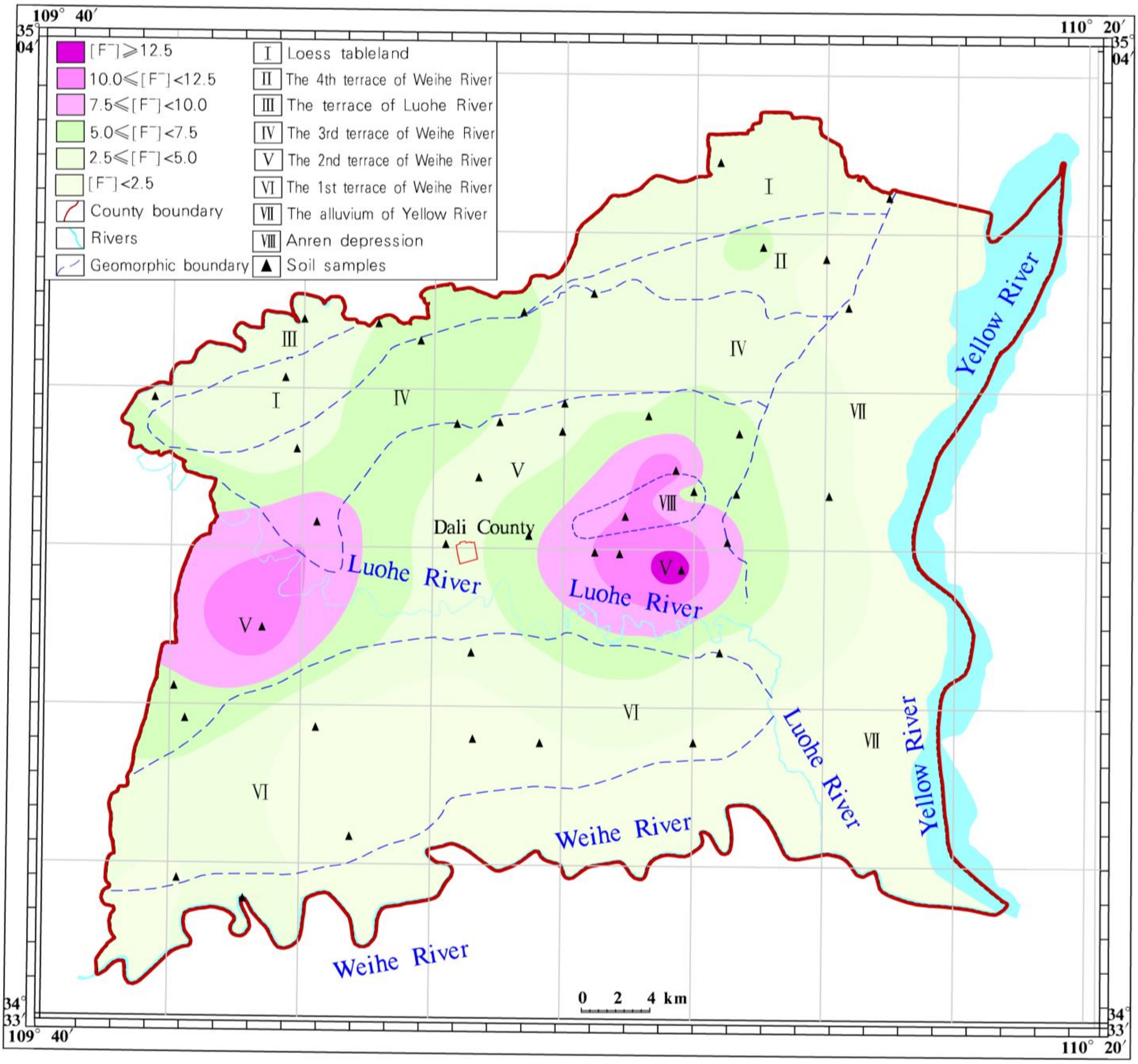

Figure 4. The distribution map of the soil fluorine contents.

2.3.4. Corn Sampling and Analysis

Corn is a widely planted food crop in Dali County, so this study took corn as the main food chain monitoring object [28]. Corn sampling points were distributed in the farmland of each geomorphic area in the study area (Figure 5). The samples were air-dried at room temperature and weighed to prepare 500 g sub-samples for fluorine content measurement. The fluorine contents were determined at the Xi'an Geological Survey Center of China in accordance with the National Code for Determination of fluorine in foods (GB/T 5009.18- 2003) [29], using the ISE-based method.

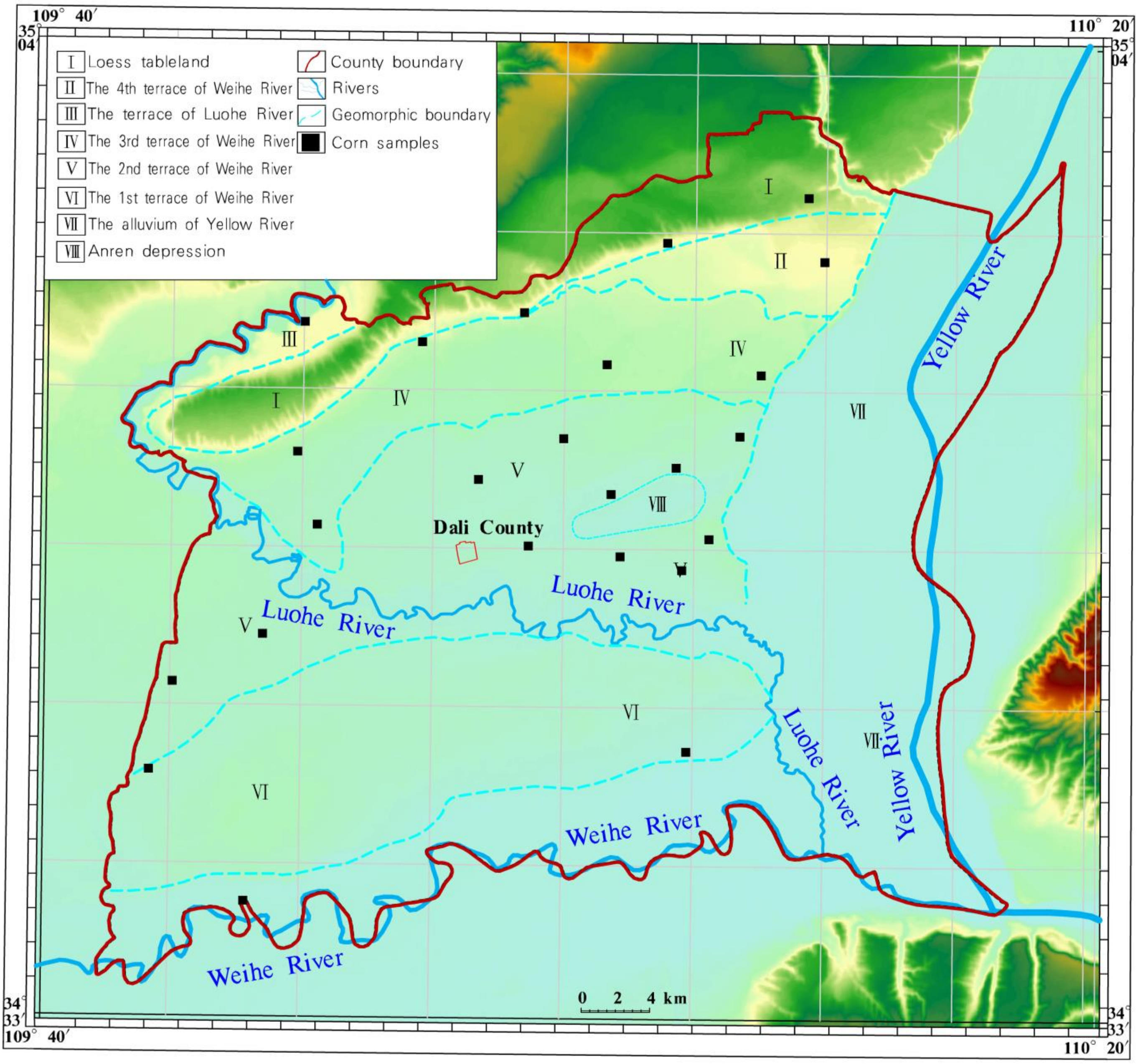

Figure 5. The distribution map of corn samples.

3. Results

3.1. Distribution of Dental Fluorosis

This research investigated the condition of dental fluorosis in seven villages selected from different geographical and geological areas, with the dental fluorosis index determined. As shown in Table 1, the higher values of dental fluorosis index 1.9 and 1.4 appeared in Lianjia village and Xinfeng village, respectively. According to China's diagnostic standard for dental fluorosis (WS/T 208-2011) [26], it should be attributed as moderately prevalent. The dental fluorosis index was both 0.7 in Fupo village and Jingzhuang village, which belonged to the mild prevalence category. The dental fluorosis index of 0.5 was observed in Caizhuang village, which is within the permitted range. The dental fluorosis index in Dongyi village and Jiaxi village were 0.2 and 0.0, respectively, which is not epidemic.

Table 1. The dental fluorosis index of the survey villages.

Villages	Number of Tested Volunteers	Number of People with Normal Test Results	Number of Suspected Cases	Number of Very Mild Cases	Number of Mild Cases	Number of Moderate Cases	Number of Severe Cases	Dental Fluorosis Index
Xinfeng	62	5	1	24	31	1	0	1.4
Caizhuang	51	29	13	2	4	3	0	0.5
Lianjia	58	0	13	13	14	12	6	1.9
Jiaxi	46	46	0	0	0	0	0	0.0
Fupo	71	29	0	35	7	0	0	0.7
Dongyi	126	108	0	17	1	0	0	0.2
Jingzhuang	115	72	0	18	12	13	0	0.7

3.2. Characteristics of Fluorine Content in Groundwater

The characteristic values of fluorine contents measured in 269 samples are shown in Table 2. The variation range of fluorine content in groundwater in Dali County is 0.01 to 11.8 mg/L. The highest fluorine content was 1180 times its lowest limit revealing a wide range of variation. The mean value was 1.91 mg/L and the median value equal to 1.71 mg/L. The fluorine contents of most groundwater samples were between 1.0 to 4.0 mg/L, accounting for 63.2% of the total samples. The fluorine contents in the groundwater of Tongguan County, which is separated by the Weihe River on the southeast side of the study area, is generally 0.3 to 0.4 mg/L and the highest value is below 0.8 mg/L [21]. The average content in this study area was more than 10 times higher than that in Tongguan County, and nearly 1.91 times higher than the limit of fluorine content in drinking water specified in the China National Standard for groundwater quality (GB/T 14848-2017) [30].

Table 2. Characteristic values of fluorine content in groundwater.

Statistical Characteristics	Content Range (mg/L)	Average (mg/L)	Mode (mg/L)	Median (mg/L)	Deviation	Fluorine Content Limits of Drinking Water (mg/L)
Character values	0.01~11.80	1.91	0.01	1.71	1.7	≤1.00

According to the quality standards for groundwater, high fluoride groundwater is defined when the fluorine level in water is more than 1.0 mg/L. Medical studies have found that the fluorine content in drinking water is closely related to endemic fluorosis (GB/T 17018-2011) [31], the best fluorine content in drinking water is between 0.5 and 1.0 mg/L, less than 0.5 mg/L will cause dental caries, osteoporosis and other diseases due to fluoride deficiency. Higher than 1.0 mg/L may lead to fluorosis and toxic effects in humans [18]. When the groundwater fluoride concentration is between 1.0 and 2.0 mg/L, the prevalence of dental fluorosis in the population will exceed 30%; when the groundwater fluoride concentration is between 2.0 and 4.0 mg/L, the prevalence of dental fluorosis in the population will exceed 80% (GB/T 17018-2011) [31]. On the basis of results given by analyses, the fluoride contents observed in groundwater collected in the study area were divided into five grades (Figure 3). The high fluoride groundwater analyzed in the study area accounted for 70.6%. The samples of which the fluorine content was between 2.0 and 4.0 mg/L accounted for 34.2% of the sampled water. It is evident that the high fluoride groundwater dominated the study area.

As shown in Figure 3, the spatial distribution of fluorine content in the sampled groundwater in the study area is uneven, with an overall trend of being higher in the north and lower in the south. The highest concentration occurs along the eastern area of the Weihe 3rd terrace and in the Anren depression. The areas with high fluoride in groundwater in the whole county account for 63% of the total investigated area. They are mainly distributed in the Loess Plateau, the Luohe terrace and the 2nd, 3rd, 4th terraces of the Weihe River. The area with increased high fluoride content (F > 4.0 mg/L) appears at the east end of the Weihe 2nd and 3rd terraces, and this may be related to the relatively low topography, shallow groundwater depth and poor excretion. The fluoride contents of groundwater in the 1st terrace of the Weihe River and in the Yellow River alluvium were below 1.0 mg/L, meeting the limit for safe drinking.

3.3. Characteristics of Fluorine Content in Soils

The soil within a depth of 20 cm is also named the aeration zone. It is the layer where water vapor and surface water connect and exchange with groundwater. It is a complex system consisting of soil particles, water and air, which has the ability to absorb water, maintain water and transfer water. The total fluorine content of soils globally fluctuates widely, averaging 200 mg/kg [32]. The total fluorine content in soils in China is higher than that in soils in the world [32].

The surface soil was divided into 6 grades according to the fluorine content (Figure 4): the fluorine content less than 2.5 mg/kg appeared in Weihe alluvium in the south of Dali County and the Yellow river alluvium in the east of Dali County. Fluorine content ranging from 2.5 mg/kg to 5.0 mg/kg was found in the west and east parts of the loess tableland, the 1st terrace of Weihe River, the middle part of the 2nd terrace of Weihe River, the east part of the 3rd terrace of Weihe River and most parts of the 4th terrace of Weihe River. Fluorine content ranging from 5.0 mg/kg to 7.5 mg/kg appeared in the middle part of loess tableland, east and west parts of the 2nd terrace of Weihe River, west part of the 3rd terrace of Weihe River and small parts of the 4th terrace of Weihe River. Fluorine content from 7.5 mg/kg to 10.0 mg/kg were mainly found in the west and east parts of the 2nd terrace of Weihe River.

3.4. Characteristics of Fluorine Content in Corns

Plants absorb fluoride from the soil, mainly concentrated in the root system, while it is rare in the fruit. When the water-soluble fluoride content exceeds 10 mg/kg, a large amount of fluoride will be absorbed and accumulated in crops [7]. In China, the fluorine contents of corn in fluorine rich soils (soil fluorine content > 500 mg/kg) are more than 10 mg/kg. Based on fluoride content's measurement results in corn grain crops from Dali County, it is obvious that the corn fluoride contents range generally from 4.04 to 7.72 mg/kg with a mean of 5.59 mg/kg, a median of 5.53 mg/kg and a deviation of 0.95 (Table 3). The fluorine contents in corn already were greater than the national food safety standard (GB 2762-2017) [33].

Table 3. Characteristic values of fluorine content in corn.

Statistical Characteristics	Content Range (mg/kg)	Average (mg/kg)	Mode (mg/kg)	Median (mg/kg)	Deviation	Fluorine Limit of Food (mg/kg)
Character values	4.04~7.72	5.59	6.14	5.53	0.95	≤1.50

4. Discussion

The tolerance and toxicity range of fluoride is very narrow and slight deviations from the optimum range may cause deep problems on human health [34]. Injuries to the body caused by fluorosis include the skeletal and non-skeletal systems impact. Bone tissue damage is mainly manifested by pathological changes of skeleton and teeth, skeletal fluorosis and dental fluorosis [35]. Fluorine has a strong bone-affinity and easily accumulates in bone tissue to form fluorosis, including osteomalacia, osteosclerosis, osteoporosis, ossification of surrounding soft tissues and degenerative changes of cartilage and joints [36]. The tooth is the most sensitive organ to fluoride and its enamel's development is damaged as dental fluorosis. The clinical manifestations of dental fluorosis are mainly the chalky change, staining and enamel defect. The damages due to fluoride which affect teeth are mainly destroying the normal structure of enamel. When the body intakes excessive fluoride, the enamel is destroyed, and the surface of teeth becomes uneven, loses normal gloss and calcium, resulting in solid light yellow, brown and black pigmentation. Teeth will become brittle and easy to wear and tear off. Previous studies on genotoxic and chronic oxidative damage mechanisms of fluoride to mammalian cells in vivo showed that, with increasing concentrations of NaF exposure, the rate of bone marrow hyperchromatic erythrocyte micronuclei and the rate of chromosomal aberrations in bone marrow cells were significantly higher in healthy male Swiss-albin mice, indicating that fluoride is genotoxic [37]. Dental fluorosis can be classified into three types based on the source of fluorine and the ingestion mode: water drinking pathogenic, coal-burning pollution pathogenic and tea drinking pathogenic [38]. Dali County is neither a coal mining area nor a tea production area. Further, other previous studies have discussed that the main soil contamination brought about by agricultural fertilization in Dali County is nitrate and nitrite pollution without fluoride pollution related to fertilization [19]. The fluorosis of this area is closely related to the fluorine content of drinking water [28].

4.1. Relationship between Fluoride Content in Groundwater and Dental Fluorosis Index of Residents

According to China National Standard for division of endemic fluorosis areas (GB 17018-2011) [31], the fluorine content in drinking water is closely related to endemic fluorosis. The fluorine content in drinking water is optimal between 0.5 and 1.0 mg/L [39]. If the fluorine content is lower than 0.5 mg/L, caries, osteoporosis and other diseases will occur due to lack of fluorine. If the fluorine content is greater than 1.0 mg/L, endemic fluorosis will occur and cause toxic effects on the human body. When the fluoride content is between 1.0 and 2.0 mg/L, the incidence of dental fluorosis in children is more than 30%. When the fluoride content is between 2.0 and 4.0 mg/L, the morbidity of dental fluorosis is more than 20%. When fluorine content in water is more than 4.0 mg/L, the incidence of dental fluorosis is more than 40% [40,41]. According to the relationship above, five grades were divided from the fluorine content of groundwater in the study area (Table 2). The results showed large areas of groundwater with high fluorine in the study area and the spatial distribution of groundwater fluorine contents are uneven with a general trend of high concentration in the north and low concentration in the south. The highest value (over 4.0 mg/L) appears in the east of the 3rd Weihe Terrace and the Anren depression. The population dental fluorosis index of Lianjia village and Xinfeng village in the Anren depression reached 1.9 and 1.4, respectively. The lowest value (under 0.5 mg/L) appears in the middle of the Weihe 3rd terrace and in the east of the 4th terrace of the Weihe River. The population dental fluorosis index of Jiaxi and Dongyi villages in low fluorine areas are 0.0 and 0.2, respectively. Therefore, the groundwater fluoride content in Dali County is positively correlated with the population dental fluoride index with a coefficient of 0.72 (Table 4). This phenomenon may indicate a dominant factor of groundwater fluoride contents on dental fluorosis.

Table 4. The relationship between the fluorine contents of ground water and the dental fluorosis index in the survey villages.

Villages	Xinfeng	Caizhuang	Lianjia	Jiaxi	Fupo	Dongyi	Jingzhuang
Fluorine content in ground water (mg/L)	6.00	2.76	5.14	2.26	3.20	2.76	2.10
Dental fluorosis index	1.40	0.50	1.90	0.00	0.70	0.20	0.70
Coefficient				$r^2 = 0.72$			

4.2. Relationship between Fluoride Content in Corn and Dental Fluorosis Index of Residents

Previous research have compared the enrichment index of water-soluble fluorine in soils with the enrichment index of fluorine in the crops under cultivation, and they showed the fold enrichment in crops is much lower than that in soils [42]. Therefore, it confirmed that the increase of fluoride content in irrigation water has little impact on the fluoride content of food crops, and the ability of plant grains concentrating fluoride is weak [43,44]. The grain and vegetable fluorine contents of the high fluorine background areas were all significantly higher than those of the low fluorine areas, indicating that grain and vegetable can enrich water-soluble fluorine from soil and irrigation water [45,46]. In order to reduce the risk of human fluoride exposure, irrigation of crops with high fluoride content groundwater should be avoided and crop varieties with lower fluoride concentration capacity should be extended and planted in the areas with high fluoride background. Roots, leaves and flowers of plants are organs with fast metabolism, and their fluorine content is significantly higher than that of grains and fruits [47]. Since a considerable amount of fluorine in food is absorbed by the human body, the choice of crops grown with the grains, fruits and stems with lower capacity of enriching fluorine as the main crops is beneficial for reducing the population fluorosis.

As shown in Table 4, the dental fluorosis index among the residents of the seven investigated villages in Dali County has a relatively clear positive correlation with the fluorine content of groundwater, indicating that dental fluorosis among the residents of Dali County is mainly caused by the citation of high fluoride groundwater. The fluorine contents of corn kernels were much lower compared to the groundwater fluorine contents. Meanwhile, poor correlation coefficient was displayed between the population dental fluorosis index of the seven surveyed villages and the fluorine content of the corn (Table 5). In other words, the consumption of high fluoride crops by the inhabitants of Dali County is not the main pathway causing dental fluorosis.

Table 5. The relationship between the fluorine contents of corn and the dental fluorosis index in the survey villages.

Villages	Xinfeng	Caizhuang	Lianjia	Jiaxi	Fupo	Dongyi	Jingzhuang
Fluorine content in corn mg/kg	5.71	6.72	4.14	5.60	6.71	5.58	4.86
dental fluorosis index	1.4	0.5	1.9	0.0	0.7	0.2	0.7
Coefficient				$r^2 = 0.26$			

4.3. Formation of High Fluorine Groundwater

4.3.1. Soil Mass Factors

Fluorine-rich soils are widely distributed in the Guanzhong Basin, mainly Quaternary loess and clayey soils, which contain fluorinated minerals such as mica, amphibole, tourmaline and clay minerals. In this sampling analysis, the perfluorine content in loess of the study area is 619.2 mg/kg, the water-soluble fluorine content is 7.0 mg/kg, the perfluorine content in clay is 628.6 mg/kg, and the water-soluble fluorine content is 7.15 mg/kg, which is close to that in loess. Paleozoic carbonate rocks and clastics are exposed in the north of the study area [48]. The lithologies are mainly limestone, marble, quartz sandstone and mudstone, and the fluorine content of marble, shale and mudstone are higher, followed by limestone and sandstone. The total fluorine content of marble is 540 mg/kg and the water-soluble fluorine is 7.5 mg/kg [8]. The total fluorine content of mudstone is 600–700 mg/kg and water-soluble fluorine is 6.2–12.5 mg/kg [14]. Higher fluorine content in rocks and loess is interacted with water and rock resulting in higher fluorine content in groundwater [49].

As shown in Figure 4, in the Loess tableland and Weihe River 4th terrace in the north of the study area, the soil is mainly loess or loess-like clay, with well-developed vertical joints and pores. Vertical infiltration of atmospheric precipitation results in strong leaching. The average soluble fluorine content in the aeration zone is 4.71 mg/L of the whole county. The area has a large hydraulic slope, good lateral runoff conditions and a deep diving water level. Fluorine found in groundwater may come from the dissolution and leaching of fluorinated minerals at the water-rock interface. The contents of soluble fluorine observed in soils from the 2nd and 3rd terraces of Weihe River averaged 7.79 mg/L and 6.27 mg/L, respectively. The lateral leaching process may be more obvious and the groundwater fluorine content may be increased due to the slower hydraulic gradient. Meanwhile, the shallow hosted groundwater exacerbates evaporation and concentration. Fluorine found in groundwater rises along the soil capillary pores and is adsorbed by clay minerals, undergoing a leaching which increases fluoride content in the groundwater. In case of rainfall infiltration, fluoride minerals in the water rock interface leach into the groundwater, and fluoride content in the groundwater accumulates. The soluble fluoride content in the soil of the Anren depression reached the maximum in the study area, due to the concentration and evaporation of groundwater. The fluorine content in the soil of the Yellow River sediment is very low since fluorine is carried away by water.

Contents-soluble fluoride observed in the soil water from the Weihe 2nd and 3rd terraces and the Anren depression were higher than that in the Loess tableland, the Weihe 1st terrace and the Yellow River alluvium. Therefore, the soil fluorine contents have to do with the type of landforms. Generally speaking, highly porous soils with strong leaching and migration are low in fluorine content due to salt dissolution by water. The finer the particles, the higher the fluorine contents in low-pore soil. This is due to the large pore surface area and strong ion exchange capacity of fine particles, and the fact that charged fine particles have strong adsorption capacity for various ions.

4.3.2. Terrain Factors

Topography plays a certain role in the enrichment of fluorine in groundwater [9]. The terrain of the study area is high in the north and low in the south, with an elevation difference of 142 m [19]. The loess tableland is located in the northern part with flat top surface and developed gullies along the tableland. The terraces of Weihe River are located on the south side of the loess tableland. The elevation difference between the terraces are small and the interior terrain of the terraces are flat with many depressions. For example, the Anren depression in the 2nd terrace of the Weihe River is about 3 to 5 km wide from north to south and about 15 km long from east to west. The groundwater depth in the depression is shallow, and some areas are even exposed on the surface to form a Brine Lake with a salinity of 28.59 mg/L and fluorine content of 1.61 mg/L [19]. The elevation of the Anren depression is even lower than that of Luohe River. Comparing the average fluorine contents of groundwater in the eight geomorphic types (Figure 3), the fluorine content of groundwater in the 2nd terrace of Weihe River is the highest with an average of 2.6 mg/L. The average fluorine contents in the Yellow River alluvium and the 1st terrace of Weihe River are less than 0.8 mg/L, meeting the China National Standard of drinking water (GB 5749–2022) [50]. The fluorine contents of groundwater in the other five geomorphic types are less than the 2nd terrace of Weihe River, but the average fluorine contents are over 2 mg/L and beyond the average fluorine content of groundwater in the whole county (1.91 mg/L) [19]. Therefore, obvious differences of the fluorine content in groundwater exist among different geomorphic types. The order of the fluorine contents from high to low is as follows: the 2nd terrace of Weihe River > the loess tableland > the 3rd terrace of Weihe River > the 4th terrace of Weihe River > the terrace of Luohe River > Yellow River alluvium > the 1st terrace of Weihe River. The landform plays a certain control effect on the fluorine content in groundwater.

4.3.3. Hydrogeological Conditions

The migration and aggregation of fluorine in groundwater are usually controlled by the characteristics of groundwater movement [48]. The high fluoride aquifer in Dali

County is mainly Quaternary pore fissure phreatic water [14]. The lithotypes containing high fluoride are loess, subclay, powdered sand, fine sand and grittite [19]. The 2nd and 3rd terraces of Weihe River are low in elevation and shallow in groundwater depth. The aquifers have a complex structure with fine grains and poor water permeability [8]. According to the pumping test in previous study, the diving hydraulic slope is in the range of 5–13% and the permeability coefficient is in the range of 0.04–1.32 m/d [21]. The hydrodynamic conditions in this area are weak, the runoff is slow and the seepage age lasts from several hundred to thousand years [21]. The alternating process of water cycle in the aquifer is extremely slow and the dissolution time of water and rock is long. The concentration of water-soluble salts is high, the pH value is generally greater than 7.3 and the alkaline water is conducive to the enrichment of fluorine. As is shown in Table 4, the groundwater fluoride contents in Xinfeng and Lianjia villages are 6.0 and 5.14 mg/L, and the dental fluorosis indexes are 1.4 and 1.9, respectively, making them high incidence areas of desfluorosis.

5. Conclusions

In this study, dental fluorosis was investigated in seven villages from Dali County, Shaanxi Province, northwest China. Lianjia village near the Anren depression showed the highest dental fluorosis index of 1.9 and the lowest dental fluorosis index of 0.0 was recorded in Jiaxi village near the Yellow River alluvium. Analyses of groundwater enabled measuring fluorine contents in groundwater ranging from 0.01 mg/L to 11.80 mg/L, with its highest value equal to 2.6 mg/L observed in the 2nd terrace of Weihe River. The lowest fluorine content (0.8 mg /L) was found in the Yellow River alluvial. As for soil samples analyses, fluorine contents ranging from 1.18 mg/kg to 13.70 mg/kg were measured, with the highest value (13.70 mg/kg) found in Xifeng village near the Anren depression. The lowest value (1.18 mg/kg) was observed in the 1st terrace of Weihe River. Measurements of fluorine contents in corn gave results ranging from 4.04 mg/kg to 7.72 mg/kg, with the highest value of 7.72 mg/kg observed in the 3rd terrace of Weihe River. The lowest value (4.04 mg/kg) was found in the 2nd terrace of Weihe River. The obtained results revealed that soil leaching was the main source of fluoride found in groundwater. Severe dental fluorosis occurred at the edges of depressions and at the junction between steep and gentle slopes. The dental fluorosis index observed in seven villages did correlate with fluorine content in corn, but positively correlated with fluorine content in groundwater. Based on the above results, two strategies are thus recommended to prevent and control dental fluorosis as well as to promote sustainable development in Dali County:

1. Change the drinking water source. The phreatic water on the northern side of the Luohe River is generally high in fluoride content with the average fluoride concentration of 2.18 mg/L [19], which exceeds the China National Standard for drinking water quality (GB 5749-2022) [50]. Low fluoride groundwater is mainly distributed on the south side of Luohe River and along the Yellow River alluvium, but the groundwater fluoride content cannot meet the China National Standard for drinking water quality (GB 5749-2022) [50]. The groundwater fluorine content in the northern loess tableland is 1.24 to 1.27 mg/L [19]. Although it exceeds the China National Standard for drinking water quality (GB 5749-2022) (<1.0 mg/L) [50,51], it is lower than most parts in Dali County [52]. At present, the water source has been supplied to the residents in the vast area north of the Luohe River [53]. However, the existing water supply capacity cannot provide enough water and many residents can only continue to use shallow well water. In response to these inadequate water resources management, the exploitation of the groundwater in the north loess tableland area and the low fluoride groundwater on the south side of the Luohe river should be properly increased, water delivery technology should be improved, and water consumption costs should be reduced, so that residents can fundamentally escape from the shortage of low fluoride drinking water.

2. Water purification. At present, a variety of water purification technologies have become increasingly mature, and the public water purifiers have gradually installed in the communities [54]. Local governments should bring in commercial capital to vigorously promote the installation and operation of public water purifiers in rural communities. In this way, drinking water and other domestic water can be separated to improve the public safety of drinking water.

Author Contributions: All authors participated in the development of this study. Conceptualization, A.Z. and H.K.; methodology, H.C.; software, M.Y.; validation, H.K.; formal analysis, M.Y.; investigation, H.C.; writing—original draft preparation, M.Y.; writing—review and editing, M.Y.; project administration, A.Z.; funding acquisition, H.K. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Social Science Foundation of Shaanxi, grant number 2021D068, the China Geological Survey Foundation, grant number DD20221774, the National Natural Science Foundation of China, grant number 42272342 and the Natural Science Basic Research Program of Shaanxi, program number 2021JM-350.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Raw data are available upon reasonable request addressed to the corresponding author.

Acknowledgments: We are thankful to the Key Laboratory for Geohazards in Loess Areas, the Chinese Ministry of Natural Resources, and the Xi'an Geological Survey Center of China Geological Survey for their contributions to this research. The authors would like to thank the reviewers and editors for their very helpful and constructive reviews of this manuscript.

Conflicts of Interest: The authors declare no conflict of interest.

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