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ORIGINAL PAPER

Hydrogeochemical Characteristics and Health Effects of Iodine in Groundwater in Wei River Basin

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Abstract

Existing studies show that drinking high-iodine or low-iodine groundwater on a long-term basis may cause goiter and other health problems. However, currently there is a lack of systematic research on the distribution, formation and health effects of iodine in groundwater at the basin scale. Taking the Wei River Basin in the Loess Plateau as a typical study area, this paper used hydrogeological surveys, sample collection, multivariate statistical analysis, health effect evaluations, and other methods, and found that iodine content in groundwater in 60.3% of the region poses potential risks to human health. Groundwater recharge areas were iodine-deficient, mainly located on the upstream Loess Plateau of the Wei River Basin in the vicinity of the Liupan Mountain watershed, and at the piedmont of the Qinling Mountains. Groundwater was low mineralization, neutral, and low-F bicarbonate, and was controlled by the weathering and dissolution of silicate minerals and evaporites, the dissolution and precipitation of carbonates, and active water circulation conditions. Iodine-deficient endemic goiter had a potential incidence of 5–38% in these areas. Groundwater runoff areas had suitable groundwater iodine content, and the groundwater hydrochemical type was dominated by HCO_3–Ca, HCO_3–Na, and $HCO_3{\cdot}SO_4$–Na type water. The mineralization degree was modest and the I^- content distribution of suitable iodine content areas was controlled by relatively active water circulation conditions. Groundwater discharge areas were high-iodine groundwater areas, where the groundwater hydrochemical type was dominated by meta-alkaline and alkaline $HCO_3{\cdot}SO_4$, $SO_4{\cdot}Cl$, and $Cl{\cdot}SO_4$ type water, and controlled by the evaporation–concentration of shallow groundwater, the biodegradation of enriched organic matter, and the competitive adsorption of HCO_3^- and I^-. Here iodine-excess endemic goiter had a potential incidence of 5–100%. Considering the results from the study area, this paper recommends that the groundwater iodine safety range for endemic goiter be set to 10.0–300.0 μg/L.

Keywords Iodine · Endemic goiter · Health effects · Hydrogeochemistry · The Wei River Basin

Introduction

Iodine is an essential trace element for human growth and development and a principal component in the synthesis of thyroid hormones. According to existing research, the effects of iodine on human health are bilateral, that is, both a deficiency and an excess of iodine can cause diseases, the most common of which is goiter (commonly known as "big neck disease") (Chen et al. 2004; Laurberg et al. 2010; Andersen et al. 2012). Iodine deficiency is a problem in 130 countries worldwide, and about 1/3 of the population live in iodine-deficient areas and is exposed to goiter risk (Hu et al. 2005; de Benoist et al. 2008; Xue et al. 2019a, b). Meanwhile, goiter caused by high iodine has also been reported in countries such as Japan, Argentina, and Denmark (Harada et al. 1994; Andersen et al. 2002; Watts et al. 2010; Voutchkova et al. 2014a, b). Water-sourced iodine is absorbed by the

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human body and goiter caused by high or low water-sourced iodine has become a worldwide public health problem.

Groundwater is one of the main water supply sources and its quality directly affects the health of residents (Li and Wu 2019a, b; Li et al. 2019a; He and Wu 2019; He et al. 2019a, b; de Moraes et al. 2019). The iodine content in groundwater is closely related to iodine deficiency disorders (IDDs) and iodine-excess disorders (IEDs). Goiter caused by groundwater iodine deficiency mainly occurs in mountainous and hilly regions (Lin et al. 1991, 2004; Cao et al. 2004; Ren et al. 2008). High-iodine groundwater has also been reported around the world. For instance, in Skagen, Denmark, the iodine content in drinking water can be as high as 140 μ/L (Andersen et al. 2012); in La Pampa of Argentina, it ranges from 52 to 395 μg/L (Watts et al. 2010); in the Horonobe area, northern Hokkaido, Japan, it is up to 270 m mol/L (Togo et al. 2016). In China, groundwater iodine deficiency is a prominent problem. With the implementation of the "universal salt iodization (USI)" program and more research on IDD prevention and control, iodine deficiency has been well controlled. However, since the discovery of iodine-excess goiter in Hebei Province in the late 1970s, many similar cases have been reported in other regions (Yu et al. 1980; Farebrother et al. 2018). So far, high-iodine groundwater has been discovered in the Datong Basin, the Guanzhong Basin, the Taiyuan Basin, the Hetao Plain, the North China Plain, and many other regions (Li et al. 2017a; Duan et al. 2016; Tang et al. 2013; Xu et al. 2013).

According to existing research, high-iodine water is mainly distributed in arid and semi-arid regions, alluvial–proluvial plains, and coastal regions (basically following a platy distribution, and supplemented by a small platy distribution and focal distribution) (Shen et al. 2007). The formation, migration, and enrichment of iodine in groundwater are mainly controlled by factors such as temperature, pH value, redox potential, and organic matter content (Otosaka et al. 2011; Li et al. 2013, 2014a; Lemieux et al. 2019; Xue et al. 2019a, b). Shallow high-iodine groundwater is mainly controlled by evaporation–concentration, while deep high-iodine groundwater is restricted by organic matter-rich reducing environments (Xu et al. 2013; Voutchkova et al. 2014a, b; Lu et al. 2015; Pi et al. 2015; Qian et al. 2017). Agricultural irrigation is also a key factor in the formation of shallow high-iodine groundwater (Li et al. 2016a).

However, at the large basin scale, differences in geomorphology, hydrogeological conditions, and climatic conditions have complicated the hydrogeochemical characteristics of iodine in groundwater, making it difficult to interpret the distribution and health effects of iodine. Some scholars have explored the distribution and factors controlling iodine in groundwater in the North China Plain, but they mainly focused on high-iodine groundwater (Zhang et al. 2013; Gao et al. 2014; Li et al. 2017a; Xue et al. 2018) and rarely considered the formation and health effects of iodine deficiencies or suitable iodine content in groundwater. For this reason, the Wei River Basin (the largest basin in the Loess Plateau in China) was selected as the study area. This paper studied the hydrogeochemical characteristics of iodine in groundwater, extracted the controlling factors and formation of iodine-deficient, enriched, and suitable content areas, and evaluated the health effects of iodine in groundwater to provide a scientific basis for the development and utilization of groundwater at the basin scale while protecting human health.

Study Area

Originating in the Niaoshushan Mountains, in Weiyuan County of Gansu Province, the Wei River is the largest tributary of the Yellow River (Li et al. 2014b). It runs through the southeast of the Loess Plateau and has a total length of 818 km. The Wei River Basin (104°–110° 20′ E, 33° 50′–37° 18′ N) extends over an area of 135,000 km^2 (Fig. 1). This is an important area for the development of the new Silk Road under the Belt and Road Initiative (Li et al. 2015, 2017b).

The water system of the Wei River Basin has an asymmetric distribution. There are more branches on the south bank, which originated from the Qinling Mountains and are characterized by clean water, short headwaters, and fast flow. Aside from having a larger basin area, there are fewer branches on the north bank, which originated from the Loess Plateau and hilly regions and have high sediment concentrations (Li and Qian 2018a, b). Within a semi-humid to semi-arid warm temperate climate zone, the climate type here is a continental monsoon climate with mean annual temperatures ranging from 7.8 to 13.5 °C, mean annual precipitation of 350–700 mm (mainly in the south and west), and mean annual evaporation of 1000–2000 mm. The study area is high in the west and low in the east, occupied by the Loess Plateau in the north and the Qinling Mountains in the south.

After considering topography and geomorphology, surface and underground watersheds, and other conditions, this paper divided the groundwater system of the Wei River Basin as follows: Western Gansu Loess Plateau Subsystem (I), Eastern Gansu Loess Plateau Subsystem (II), Northern Shaanxi Loess Plateau Subsystem (III), Guanzhong Basin Subsystem (IV), and North Qinling Mountains Piedmont Subsystem (V) (Sun et al. 2016).

Western Gansu Loess Plateau Subsystem (I) is located upstream of the Wei River Basin and its main aquifers are alluvial–pluvial rubble, gravel layers, and loess layers. Atmospheric precipitation is the primary recharge source for groundwater in this subsystem, and groundwater is mainly discharged to the Wei River and the gullies on both sides of the valley.

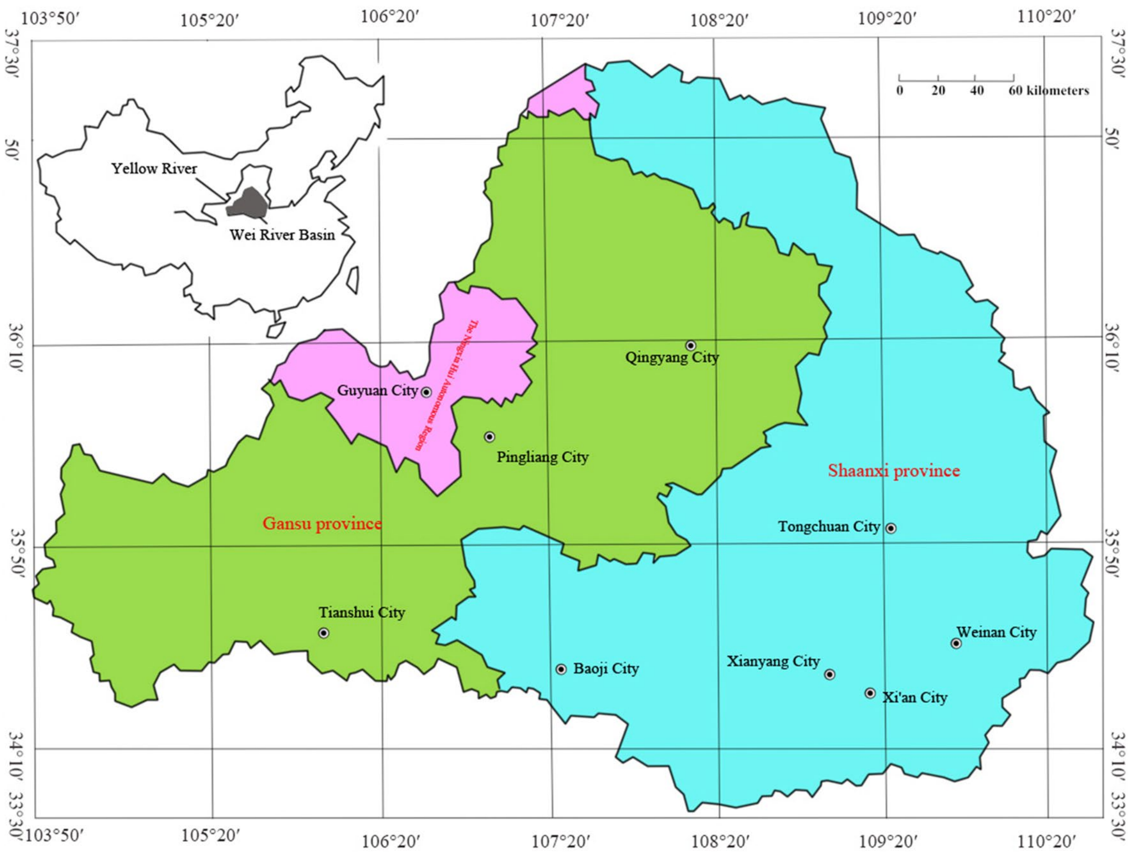

Fig. 1 Geographical location map of Wei River Basin

Eastern Gansu Loess Plateau Subsystem (II) is located midstream of the Wei River Basin and its main aquifers are alluvial sand and pluvial sand layers distributed zonally in the Jinghe River Valley and piedmont alluvial–pluvial fans. Groundwater here is mainly laterally recharged by atmospheric precipitation and river water, then discharged to gullies in the form of springs.

Northern Shaanxi Loess Plateau Subsystem (III) is dominated by the Loess Plateau and its terrain is high in the northwest and low in the southeast. Groundwater types include fissure water in clastic rocks, pore water in loose rocks, and pore-fissure water widely distributed within loess layers (Li and Qian 2018a, b). Atmospheric precipitation is the primary recharge source for groundwater in this subsystem, which is mainly discharged to gullies in the form of springs.

Guanzhong Basin Subsystem (IV) is located in the west part of the Fen-Wei fault-depression zone and its aquifers are alluvial–proluvial coarse rubble and gravel layers. Groundwater here is mainly recharged by atmospheric precipitation and discharged through evaporation, horizontal discharge to rivers, springs, and artificial exploitation (Wang et al. 2018; Li et al. 2016b, c, 2019a). North Qinling Mountains Piedmont Subsystem (V) is mainly weathering fissure water in a near-surface distribution, with poor water yield.

Materials and Methods

Water samples were collected twice from the study area. In the first campaign (2011–2013), we collected 404 water samples, including 365 groundwater samples, 16 spring water samples, eight cellar water samples, and 15 river water samples. In the second campaign (2017), we collected 24 supplementary water samples from high-iodine groundwater regions, including 21 groundwater samples, two river water samples, and one canal water sample.

In this study, all groundwater samples were collected from domestic drinking water wells approximately 30 min after pumping and river water samples were collected from the main stream and branches of the Wei River. GPS was used to record the geographic coordinates and groundwater burial depth, and a portable multiparameter water quality monitor was used to test T, pH, Ec, and Eh on site. When water samples were to be used for major element and trace element analysis, 500 mL polyethylene bottles were used for sampling. During collection, water samples were rinsed 2–3 times and the bottles were filled with water. After sampling, a simple filter unit with a 0.45 µm filter membrane was used for filtration and removal of suspended impurities. Three water sample bottles were collected from each sampling point. Several drops of HNO_3 were added to one bottle until the pH was less than 2, which was used for testing cations. The other two bottles were used to test anions and other trace elements. All water samples were stored at a low temperature in an incubator.

K^+ and Na^+ were tested using flame atomic absorption spectrophotometry (WFX-110B). SO_4^{2-}, Cl^-, NO_3^-, NO_2^-, F^-, and I^- were tested using ion chromatography (HLC-601). Ca^{2+} and Mg^{2+} were tested using EDTA titration. HCO_3^- and CO_3^{2-} were tested using acid–base titration. Al, As, Cr, Se, Mn, Hg, and other trace elements were tested using ICP-MS. During sample testing, duplicate analyses were conducted on 5% of the samples for quality control. The error was < 5% for all duplicates.

Results and Discussion

Iodine Ion Content in Groundwater

The statistical results of hydrogeochemical composition indexes of shallow groundwater in Wei River Basin are shown in Table 1. Iodine in groundwater is found in three main forms, that is, IO_3^-, I^-, and organic iodine. In northern China, the dominant form is I^- (Xue et al. 2018). According to the results of the 386 groundwater samples collected from the Wei River Basin, iodine ion content in groundwater varied between 2 and 28,620 µg/L, with a mean of 764.17 µg/L, a median of 91.0 µg/L, and a variation coefficient (C_v) of 3.73. In the study area, iodine ion content in river water varied between 2 and 420 µg/L, with a mean of 110.0 µg/L, a median of 50.0 µg/L, and a variation coefficient (C_v) of 1.28. This suggests that the mean iodine ion content in river water was less than that in groundwater. From the perspective of water supply, river water can be used as a drinking water source in high-iodine groundwater areas.

Table 1 Hydrogeochemical indexes of shallow groundwater in Wei River Basin

Indexes	Unit	Minimum value	Maximum value	Average value	Standard deviation	Coefficient of variation
Groundwater depth	m	1.11	180	27.67	34.85	1.259
Temperature	°C	11.3	20.3	14.99	2.04	0.136
pH		6.51	9.67	7.78	0.61	0.078
TDS	mg/L	88.02	22,490	1100.99	1825.6	1.658
TH	mg/L	55.84	3822	477.61	445.64	0.933
Soluble SiO_2	mg/L	0.04	31.12	14.52	4.67	0.322
COD	mg/L	0.02	117.60	1.58	5.928	3.752
CO_2	mg/L	0.00	43.50	11.42	7.52	0.658
K^+	mg/L	0.17	88.17	3.33	6.58	1.976
Na^+	mg/L	2.26	5724.8	207.78	490.74	2.362
Ca^{2+}	mg/L	5.46	385.37	86.16	60.75	0.705
Mg^{2+}	mg/L	1.51	800.18	63.74	86.80	1.362
Cl^-	mg/L	3.23	5398.25	171.29	434.24	2.535
SO_4^{2-}	mg/L	2.80	9182.50	316.65	799.92	2.526
HCO_3^-	mg/L	18.42	1140.36	419.85	173.31	0.413
NO_3^-	mg/L	< 0.03	612	36.14	65.49	1.812
F^-	mg/L	0.01	4.90	0.62	0.655	1.056
As	µg/L	< 0.04	24.94	1.672	2.476	1.481
Hg	µg/L	< 0.01	0.817	0.245	0.224	0.914
Cr	mg/L	< 0.004	0.373	0.034	0.054	1.588
I^-	µg/L	2	28,620	764.17	2850.35	3.73
Mn	mg/L	< 0.001	1.39	0.432	0.168	0.389
Se	µg/L	< 0.04	0.72	0.14	0.12	0.857
Al	mg/L	< 0.02	2.54	0.180	0.307	1.706
Fe	mg/L	< 0.002	2.706	0.060	0.186	3.1

According to the national standards put forward by the Chinese Ministry of Health (GB/T 19380-2003, and GB 16005-2009), iodine content in groundwater in the study area was divided into five intervals. Table 2 presents the percentages of groundwater samples at different intervals based on testing results. As can be seen in Table 1, 22.2% of groundwater samples had an iodine content of less than 10 μg/L and were termed iodine-deficient, whereas 38.1% of groundwater samples had an iodine content of above 150 μg/L and were termed high-iodine water that may induce goiter. This suggests that iodine content in groundwater poses potential risks to human health in about 60.3% of the Wei River Basin.

Table 2 Statistics of percentages of groundwater samples with different iodine content intervals

Iodine concentration (μg/L)	< 10	10–150	150–300	300–1000	> 1000
Percentage (%)	22.2	39.7	12.1	15.0	11.0

Spatial Distribution of Iodine in Groundwater

The iodine spatial distribution map of the groundwater in the Wei River Basin (Fig. 2) was drawn using MAPGIS. As can be seen from Fig. 2, that iodine content in groundwater in the study area had clear zonality in the horizontal direction. From the piedmont (the Loess Plateau) to the center of the Guanzhong Basin, and from upstream to downstream Wei River, iodine content in groundwater gradually declined, which was basically consistent with the groundwater flow direction.

The zoning map of iodine content (Fig. 2) shows that iodine content in groundwater was generally low (< 10 μg/L) upstream of the Wei River and in the piedmont alluvial fan of the Qinling Mountains. These regions are the main recharge areas for groundwater in the Wei River Basin where groundwater runoff was relatively smooth and 95% of groundwater had a TDS value of less than 1.0 g/L. Suitable groundwater iodine content areas with groundwater iodine contents of 10~150 μg/L were mainly distributed in Eastern Gansu Loess Plateau Subsystem and over the majority of Northern Shaanxi Loess Plateau Subsystem. The suitable groundwater iodine content areas were also found in the loess tableland,

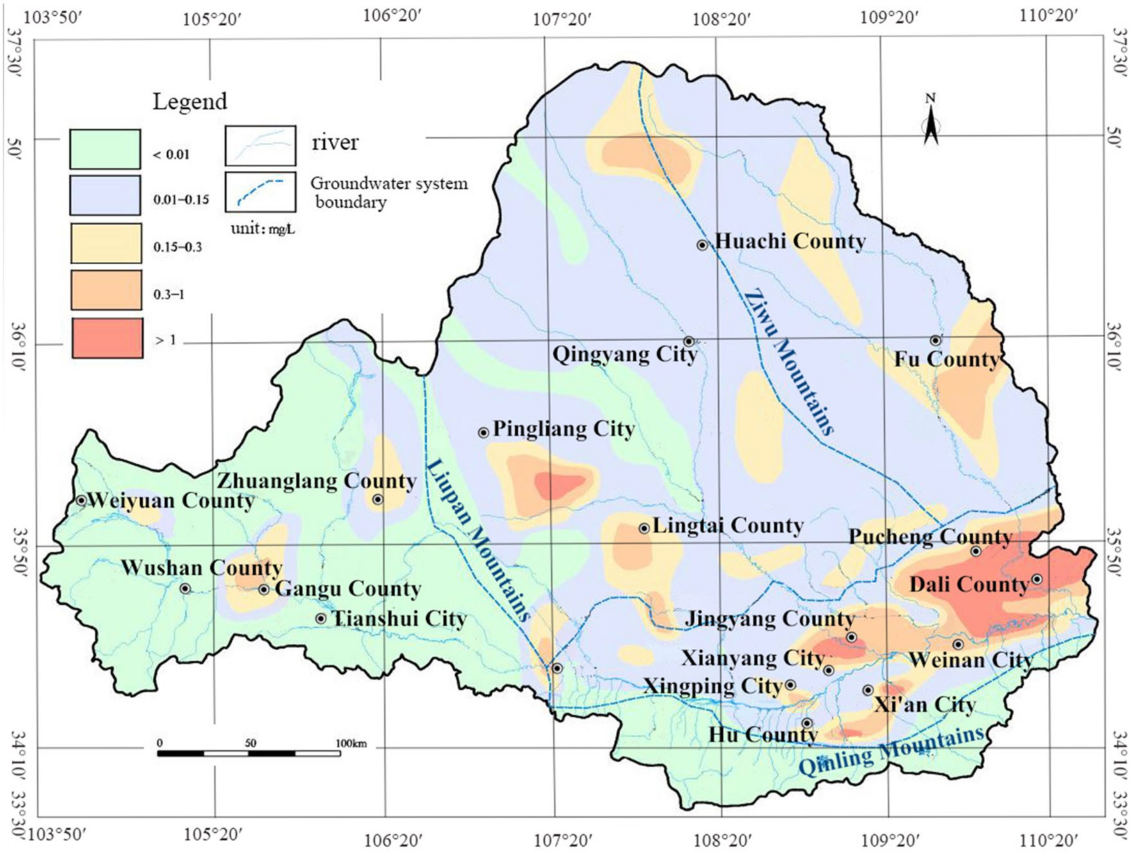

Fig. 2 Zoning map of iodine contents of groundwater in Wei River Basin

valley terrace, and some alluvial-proluvial fans to the west of the Jing River in the Guanzhong Basin Subsystem. These regions are groundwater runoff areas, where runoff was relatively smooth and 91% of groundwater had a TDS value of less than 1.0 g/L. High-iodine areas (not iodine-excess endemic goiter areas) with groundwater iodine contents of 150–300 μg/L were mainly distributed in the alluvial plains of the Guanzhong Basin, such as Xi'an, Xianyang, Xingping, Weinan, Huaxian County, and Huayin, and also scattered in Gangu County, Zhuanglang County, Pingliang, Lingtai County, and Huanxian County of Gansu Province. In these regions, 95% of groundwater had a TDS value of less than 1.5 g/L. Iodine-excess endemic goiter areas with groundwater iodine contents of 300–1000 μg/L were mainly distributed in the regions to the north of the Wei River and to the east of Liquan County in the Guanzhong Basin, partially distributed in the Wei River terrace of Weinan, Huaxian County, and Huayin and in some parts of Huxian County, and also scattered in Gangu County, Zhuanglang County, Pingliang, Lingtai County, and Huanxian County of Gansu Province. Ultra-iodine-excess endemic goiter areas with groundwater iodine contents above 1000 μg/L were mainly distributed in the majority of Pucheng County and Dali County, in Huaxian County in the Guanzhong Basin, and also scattered in Pingliang of Gansu Province and Jingyang County of Shaanxi Province. High-iodine groundwater areas were groundwater runoff discharge areas or groundwater discharge areas, where runoff is stagnant, groundwater movement shifts from horizontal to vertical, and evaporation is intense.

The environmental conditions of aquifers at different depths influenced the iodine content and form in groundwater. Given that the groundwater samples used in this study were from domestic drinking water wells, the environmental conditions of groundwater had already been disturbed by groundwater exploitation. In this case, changes in iodine content in groundwater would not fully reflect the natural aquifer conditions, but they can provide some valuable information for discussion about iodine content change in groundwater at different depths. As can be seen from Fig. 3, in aquifers with a burial depth of less than 100 m, iodine-deficient groundwater, medium-iodine groundwater, and high-iodine groundwater were detected. In deep aquifers with a burial depth greater than 100 m, medium-iodine groundwater and high-iodine groundwater coexisted, but iodine content in high-iodine groundwater was clearly lower than in shallow groundwater. This finding was consistent with the vertical distribution characteristics of iodine content in groundwater in the Datong, Taiyuan, and Guanzhong Basins, but differed greatly from iodine content distributions in groundwater in the North China Plain (Tang et al. 2013; Duan et al. 2016; Li et al. 2017a). This is mainly because the climatic and geographic conditions of the Wei River basin are similar to the Datong, Taiyuan, and Guanzhong Basins, while the North China Plain has a warm temperate humid monsoon climate and a smaller topographic slope.

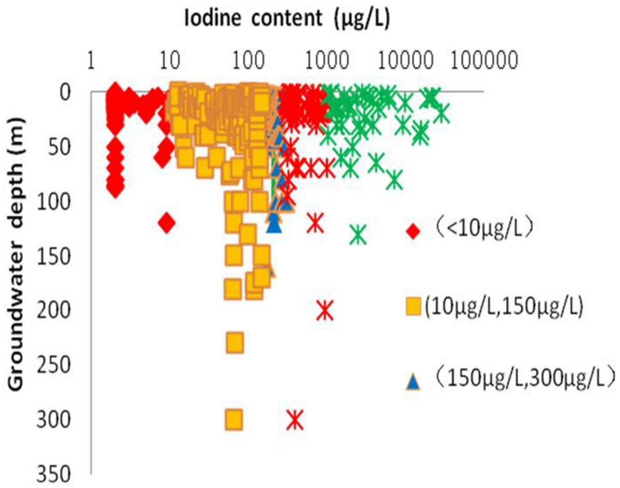

Fig. 3 Distribution of iodine content in groundwater in depth direction

Hydrochemical Characteristics of Iodine in Groundwater

Based on the above intervals of iodine content in groundwater, the hydrochemical Piper trilinear nomographs of different iodine content in groundwater were plotted (Fig. 4). As can be seen from Fig. 4, in iodine-deficient areas with groundwater iodine contents of less than 10 μg/L, cations were dominated by Ca^{2+}, anions were dominated by HCO_3^-, and hydrochemical type was dominated by HCO_3–Ca and HCO_3–Ca·Mg type water (Fig. 4a). In suitable groundwater iodine content areas with iodine contents of 10–150 μg/L, cations were dominated by Ca^{2+} and Na^+, anions were dominated by HCO_3^- and SO_4^{2-}, and hydrochemical type was relatively complicated and dominated by HCO_3–Ca, HCO_3–Na, and HCO_3·SO_4–Na type water (Fig. 4b). In high-iodine areas (not iodine-excess endemic goiter areas) with iodine contents of 150–300 μg/L, cations were dominated by Ca^{2+} and Na^+, anions were dominated by HCO_3^-, and hydrochemical type was relatively singular and dominated by HCO_3–Ca and HCO_3–Na type water (Fig. 4c). In iodine-excess endemic goiter areas with iodine contents of 300–1000 μg/L, cations were dominated by Ca^{2+} and Na^+, anions were dominated by HCO_3^- and SO_4^{2-}, and hydrochemical type was complicated and dominated by HCO_3–Ca, HCO_3–Na, HCO_3·SO_4–Ca, HCO_3·SO_4–Na, SO_4·Cl–Na, and SO_4–Na type water (Fig. 4d). In ultra-high-iodine areas with iodine contents above 1000 μg/L, cations were dominated by Na^+, anions were dominated by SO_4^{2-} and HCO_3^-, and hydrochemical type was dominated by SO_4–Na, HCO_3·SO_4–Na,

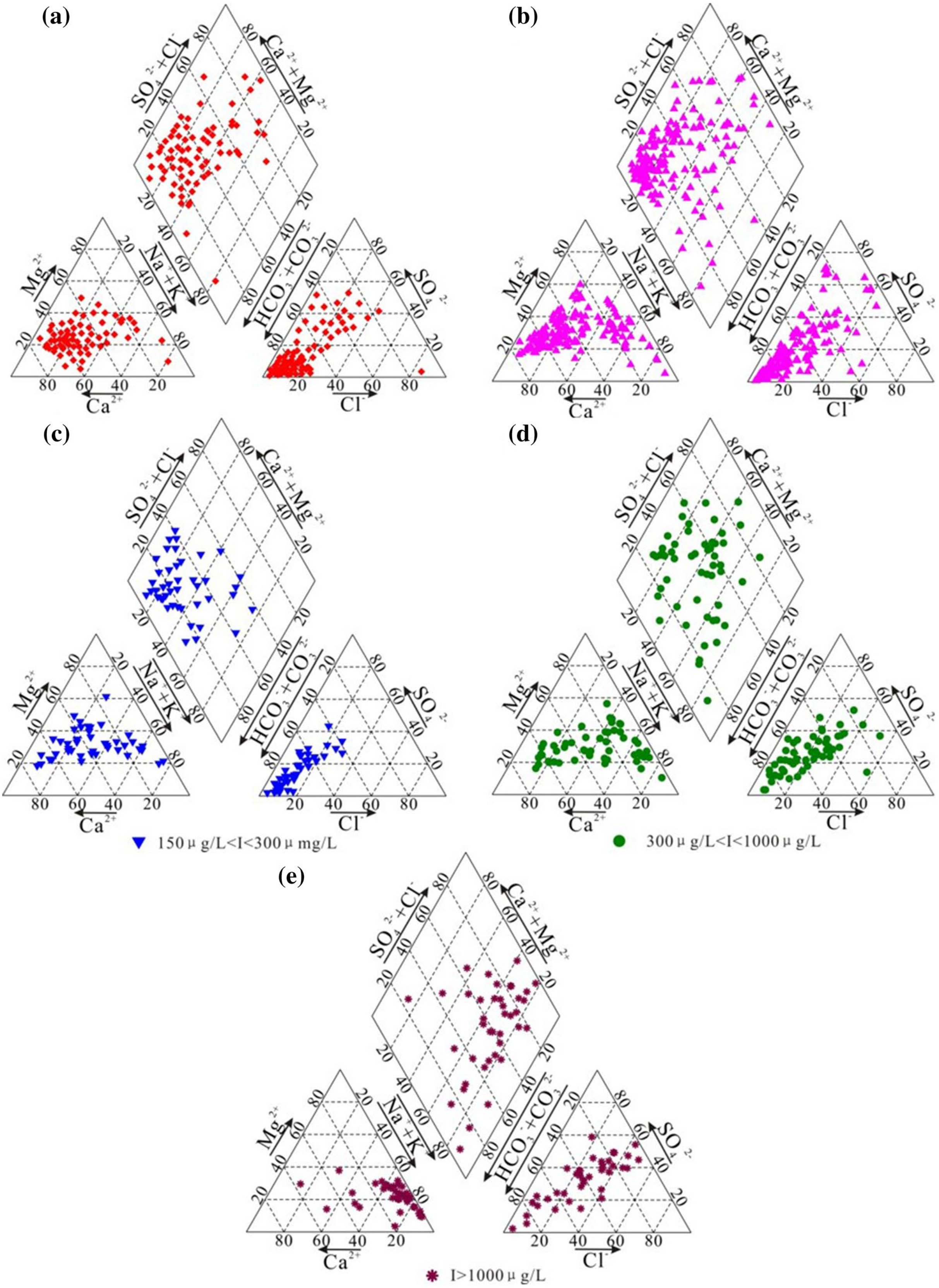

Fig. 4 Chemical types of iodine contents in groundwater

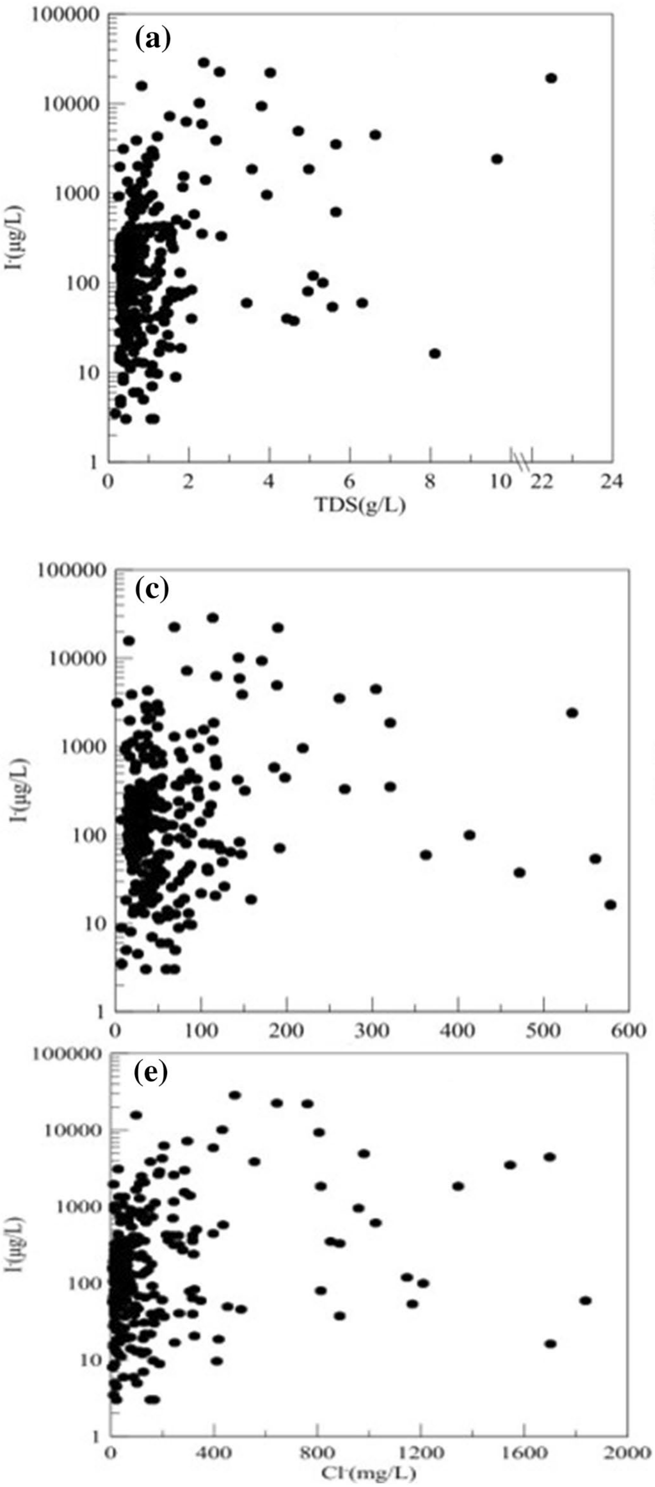

Fig. 5 Relationship between iodine and TDS and the ions

and SO_4·HCO_3–Na type water (Fig. 4e). In summary, the hydrochemical evolution of different iodine content intervals in the study area were consistent with the hydrochemical evolution of groundwater (Liu et al. 2018; Xu et al. 2019), as well as iodine in groundwater in other regions (Xue et al. 2018).

There was a clear positive correlation between iodine content and HCO_3^-, and iodine content increased with increasing HCO_3^-. However, iodine content had no clear correlation with TDS, Na^+, Mg^{2+}, SO_4^{2-}, or Cl^- (Fig. 5). Average local hydrochemical component values (Table 3) showed that as iodine content increased, TDS, Na^+, Mg^{2+}, SO_4^{2-}, Cl^-, and F^- in groundwater also increased. That is to say, iodine content in groundwater was closely related to pH, mainly occurred in meta-alkaline environments, and had a strong positive correlation with HCO_3^-. Existing studies indicate that as pH increases, iron and clay minerals produce more negative charges that reduce the adsorption of I^- or the competitive adsorption of HCO_3^-, OH^-, and I^- caused by high HCO_3^-, consequently enhancing the desorption of iodine (Dai et al. 2009; Nagata et al. 2009). This indicates that, in a high-pH environment, the iodine adsorbed by sediments is released to groundwater.

Redox potential (Eh) is a sensitive groundwater quality index that controls the speciation, migration, and transformation of iodine in groundwater (Hou et al. 2009; Li et al. 2014a). In the groundwater of the study area, Eh varied between −150 and 252 mV, and had a median of 152 mV, which indicates that aquifers were dominated by a sub-oxidation–reduction state. The statistics showed no obvious linear relationship between iodine content in groundwater and Eh. This is because Eh is a highly variable index that changes constantly in field testing, making it impossible measure accurately (Duan et al. 2016).

Differential Formation of Iodine Content in Groundwater

To discuss the formation of iodine in groundwater in the Wei River Basin, this paper selected Na^+, Ca^{2+}, Mg^{2+}, SO_4^{2-}, Cl^-, HCO_3^-, F^-, I^-, TDS, and pH as key indices (Table 4), and used Principal Component Analysis (PCA) to extract the main factors influencing iodine formation, migration, and enrichment (Li et al. 2019b; Wu et al. 2014, 2019a). The hydrogeochemical processes contributing to the formation of iodine-deficient, suitable, and high-iodine areas were discussed.

Table 3 Statistics of groundwater chemistry characteristics between different iodine content intervals

Projects	pH	TDS (g/L)	Ion content (mg/L)						
			Na^+	Ca^{2+}	Mg^{2+}	SO_4^{2-}	Cl^-	HCO_3^-	F^-
Iodine deficiency areas (lower than 10 μg/L)	7.65	0.52	55.54	76.69	35.19	68.73	29.76	374.62	0.43
Iodine appropriation areas (higher than 10 μg/L and lower than 150 μg/L)	7.86	0.51	45.37	78.01	34.59	62.27	37.32	371.45	0.36
Areas of iodine-excess and non-iodine-excess disorders (higher than 150 μg/L and lower than 300 μg/L)	7.89	0.49	52.55	58.55	33.33	52.63	35.49	395.44	0.35
Areas of iodine-excess disorders (higher than 300 μg/L and lower than 1000 μg/L)	7.65	0.80	147.5	85.05	46.6	181.8	92.76	484.35	0.39
Iodine-ultra-excess areas (higher than 1000 μg/L)	7.83	1.18	343.1	43.81	60.02	319.0	223.8	588.13	0.81

Table 4 Factor loadings of hydrogeochemical parameters in different iodine content areas

Parameters	Iodine deficiency area			Iodine appropriation area			Iodine-excess area		
	F_1	F_2	F_3	F_1	F_2	F_3	F_1	F_2	F_3
Na^+	0.96	0.11	0.15	0.96	0.14	0.07	0.96	0.22	0.04
Ca^{2+}	0.41	−0.77	−0.07	0.57	−0.73	0.10	0.36	−0.54	0.53
Mg^{2+}	0.92	−0.15	0.19	0.92	0.01	0.08	0.92	0.10	0.20
SO_4^{2-}	0.92	−0.07	−0.18	0.97	−0.04	0.05	0.98	0.10	0.05
Cl^-	0.83	−0.14	0.14	0.97	−0.06	0.03	0.99	0.08	0.08
HCO_3^-	0.28	−0.25	0.70	0.03	0.15	0.91	0.25	0.65	0.50
F^-	0.72	0.43	0.27	0.54	0.71	0.17	0.07	0.81	−0.18
I^-	−0.03	0.20	0.73	−0.06	0.02	−0.02	0.41	0.63	−0.08
TDS	0.97	−0.20	0.07	0.99	−0.04	0.05	0.98	0.13	0.08
pH	0.12	0.86	−0.04	−0.05	0.53	−0.70	−0.01	0.11	−0.91
Eigenvalue	5.17	1.72	1.07	5.30	1.55	1.20	5.38	1.89	1.10
Variance contribution rate %	51.73	17.19	10.70	53.04	15.47	11.99	53.85	18.86	11.0
Cumulative contribution rate %	51.73	68.92	79.62	53.04	68.51	80.50	53.85	72.71	83.71

In groundwater iodine-deficient areas, there were three main controlling factors that influenced the hydrochemical composition of groundwater: factors F_1, F_2, and F_3. To be specific, the F_1 factor contributed 51.73% and was closely related to TDS, Na^+, SO_4^{2-}, Mg^{2+}, Cl^-, and F^-. F_2 contributed 17.19% and had a positive correlation with pH, but was negatively correlated with Ca^{2+}. F_3 contributed 10.70% and was positively correlated with I^- and HCO_3^-. F_1 indicated that in the study area, the weathering and dissolution of silicate minerals and evaporites had a direct influence on TDS, Na^+, SO_4^{2-}, Mg^{2+}, Cl^-, and F^- content, and controlled groundwater salinity (Li et al. 2019b). F_2 showed that the dissolution and precipitation of calcium minerals were controlled by groundwater pH, but had a low correlation with I^- (0.2). F_3 identified HCO_3^- as the primary factor controlling iodine content in groundwater. In summary, the weathering and dissolution of silicate minerals and evaporites, and the dissolution and precipitation of carbonates, were key hydrogeochemical processes influencing groundwater iodine-deficient areas. In addition, these areas were mainly distributed in the piedmont proluvial fan of the Qinling Mountains and the upstream Loess Plateau of the Wei River Basin. In these regions, the aquifers were composed of highly permeable pebble gravels, medium sand, and find sand, the hydraulic slope was relatively high, and the groundwater runoff was smooth. The easy loss of I^- to rapid groundwater flows was an important contributor to I deficiency in groundwater in these regions.

Seen from Table 3, in suitable groundwater iodine content areas, there were also three main factors that influenced the hydrochemical composition of groundwater: F_1, F_2, and F_3. F_1 contributed 53.04% and was positively correlated with TDS, SO_4^{2-}, Cl^-, Na^+ and Mg^{2+}. This indicates that, in the study area, the weathering, dissolution, and evaporation of evaporite minerals had a direct influence on of Na^+, SO_4^{2-}, Mg^{2+}, and Cl^- content and controlled groundwater TDS (Li et al. 2018a, b). F_2 contributed 15.47% and was positively correlated with F^- and pH, but negatively correlated with Ca^{2+}. This suggests that the dissolution and precipitation of fluorites were controlled by the pH and further determined the Ca^{2+} and F^- content. F_3 contributed 11.99% and was positively correlated with HCO_3^-, but negatively correlated with pH. In addition, in these regions, groundwater runoff was relatively active and the loss of iodine in groundwater was non-significant. Together, these results suggest that the distribution of iodine content in suitable groundwater iodine content areas was controlled by the dissolution of evaporites, the dissolution and precipitation of fluorites, and relatively active hydrodynamic conditions.

In groundwater discharge areas, the hydrogeochemical characteristics of iodine enrichment were related to three main controlling factors, as well. F_1 contributed 53.85% and was closely related to TDS, Na^+, Mg^{2+}, SO_4^{2-}, and Cl^-. F_2 contributed 18.86%, and was positively correlated with HCO_3^-, F^-, and I^-, but had an obvious negative correlation with Ca^{2+}. F_3 contributed 11.0% and was positively correlated with Ca^{2+} and HCO_3^-, but negatively correlated with pH. F_1 indicated that the dissolution and evaporation–concentration of evaporites had a direct influence on TDS, Na^+, SO_4^{2-}, Mg^{2+}, and Cl^- content. F_2 identified HCO_3^- as the primary factor controlling iodine content in groundwater, and F^- and I^- as coexisting elements (Pi et al. 2015). F_3 showed that the dissolution and precipitation of minerals containing carbonates were also important factors influencing the distribution of iodine in groundwater, and that pH limited the distribution. In high-iodine groundwater areas, aquifers were dominated by lacustrine, eolian, and alluvial deposits, and, in terms of lithology, were mainly composed of clay, loam, sandy loam, etc. With the disappearance of ancient lakes, biochemical actions caused enrichment of iodine in plankton, hydrophytes, and other organisms (Xue et al. 2019a, b). In an enclosed and anaerobic environment, driven by the microbial metabolism and decomposition of organic matter, the iodine adsorbed on organic matter surface was released into groundwater. The competitive adsorption of HCO_3^- and I^- in groundwater further indirectly promoted the iodine enrichment (Wang et al. 2009; Xu et al. 2013).

Relationship Between Iodine Content in Groundwater and Human Health

Because of the dual thresholds for the effects of iodine on human health, both low and high iodine can cause endemic goiter (Cao et al. 2004; Voutchkova et al. 2014a, b; Voutchkova et al. 2017). There have been lots of studies on health risk quantification of various contaminants in groundwater (Li et al. 2016d, 2019c; Wu et al. 2019b; Wu and Sun 2016; Zhang et al. 2018a, b). These studies are deterministic interpretation of the health risk. However, to accurately measure the relationship between iodine content in groundwater and the endemic goiter prevalence, Wang et al. (1983) used a parabolic equation to prepare statistics on iodine content (natural logarithm taken, x) and prevalence (y), as shown below:

$$\hat{y} = 114.23 - 37.09x + 2.92x^2 \tag{1}$$

If a 5% prevalence of endemic goiter were selected as the classification criterion for endemic areas, the most suitable range of iodine content in groundwater was determined to be 10–100 μg/L according to Formula (1). When iodine content in groundwater is less than 10 μg/L, as iodine content in drinking water drops, the prevalence would increase, so there is a negative correlation between them. When iodine content in groundwater is above 100 μg/L, as iodine content increases, the prevalence would increase, so there is a positive correlation between them.

The above method was used to obtain the relationships between iodine content in groundwater and endemic goiter prevalence for different groundwater subsystems of the Wei River Basin (Fig. 6). There was no potential for endemic goiter in the Northern Shaanxi Loess Plateau Subsystem. The Western Gansu Loess Plateau Subsystem was dominated by iodine-deficient endemic goiter, which mainly occurred in mountainous and hilly regions and at the piedmont of the West Qinling Mountains with a potential incidence of 5–30%. The Eastern Gansu Loess Plateau Subsystem was exposed to the health risks of both iodine-deficient and iodine-excess endemic goiter. Iodine-deficient endemic goiter mainly occurred in the vicinity of the Liupan Mountain watershed, with a potential incidence of 5–38%. Iodine-excess endemic goiter mainly occurred in Chongxin County of Pingliang, with a potential incidence of 5–25%. The Guanzhong Basin Subsystem was dominated by iodine-excess endemic goiter, which mainly occurred in Pucheng and Dali of the Guanzhong Basin with a potential incidence of 5–100%. The North Qinling Mountains Piedmont Subsystem faced the health risk of iodine-deficient endemic goiter, which had a potential incidence of 15–50%.

To sum up, potential iodine-deficient endemic goiter mainly occurred in groundwater recharge areas characterized by high terrain, strong hydrodynamic erosion and cutting, fine surface runoff conditions, intense groundwater circulation, dominant weathering and leaching in hydrogeochemical processes, and easy loss of iodine in groundwater. Iodine-excess endemic goiter mainly occurred in groundwater discharge areas characterized by low terrain, poor hydrodynamic conditions, shallow burial depth, strong evaporation–concentration, alkaline and reducing environments, and easy iodine enrichment. In groundwater runoff areas, iodine content in groundwater was suitable, so these were the most suitable areas for mankind.

Consensus has now been reached about the relationship between groundwater iodine and endemic goiter, as well as the upper and lower thresholds. However, due to differences in prevalence among various classification criteria for endemic areas, the safety ranges differ as well (Wang et al. 1983; Lin 1991; Farebrother et al. 2018). Based on existing research results, considering water shortages in the study area and the national standards set by the Ministry of Health for the determination and classification of iodine-excess goiter endemic areas, this paper recommends that the groundwater iodine safety range for endemic goiter should be set at 10.0–300.0 μg/L for this area.

Conclusion

In the study area, iodine content in groundwater varied between 2 and 28,620 μg/L with a mean of 764.17 μg/L. Low-iodine water ($I^- \leq 10$ μg/L) was mainly distributed in groundwater recharge areas, i.e., the upstream Loess Plateau of the Wei River Basin and the piedmont alluvial fan of the Qinling Mountains, where hydrochemical type was dominated by HCO_3–Ca and HCO_3–Ca·Mg type water. High-iodine water ($I^- > 300$ μg/L) was mainly distributed in groundwater runoff discharge areas or groundwater discharge areas, and the majority of the loess tableland and valley terrace to the east of the Qishui River in the Guanzhong Basin where hydrochemical type was dominated by HCO_3–Na, $HCO_3 \cdot SO_4$–Na, $SO_4 \cdot Cl$–Na, and SO_4–Na type water. Iodine content in groundwater posed potential risks to human health in 60.3% of regions.

The I^- content distribution of groundwater iodine-deficient areas were mainly controlled by weathering and dissolution of silicate minerals and evaporites, dissolution and precipitation of carbonates, and active water cycling conditions. Suitable groundwater iodine content areas were controlled by relatively active water cycling conditions. High-iodine groundwater areas were mainly controlled by shallow groundwater evaporation–concentration, enriched organic matter biodegradation, the competitive adsorption of HCO_3^- and I^-, and stagnant groundwater flow.

The study area was exposed to the health risks of both iodine-deficient and iodine-excess endemic goiter. Iodine-deficient endemic goiter mainly occurred in groundwater recharge areas, i.e., the upstream Loess Plateau of the Wei River Basin, around the Liupan Mountain watershed, and the piedmont of the Qinling Mountains, with potential incidence of 5–50%. Iodine-excess endemic goiter mainly occurred in groundwater discharge areas, i.e., Pucheng and Dali of the Guanzhong Basin, with potential incidence of 5–100%. Based on the above results, this paper recommends that the groundwater iodine safety range for endemic goiter should be set as 10.0–300.0 μg/L for the study area.

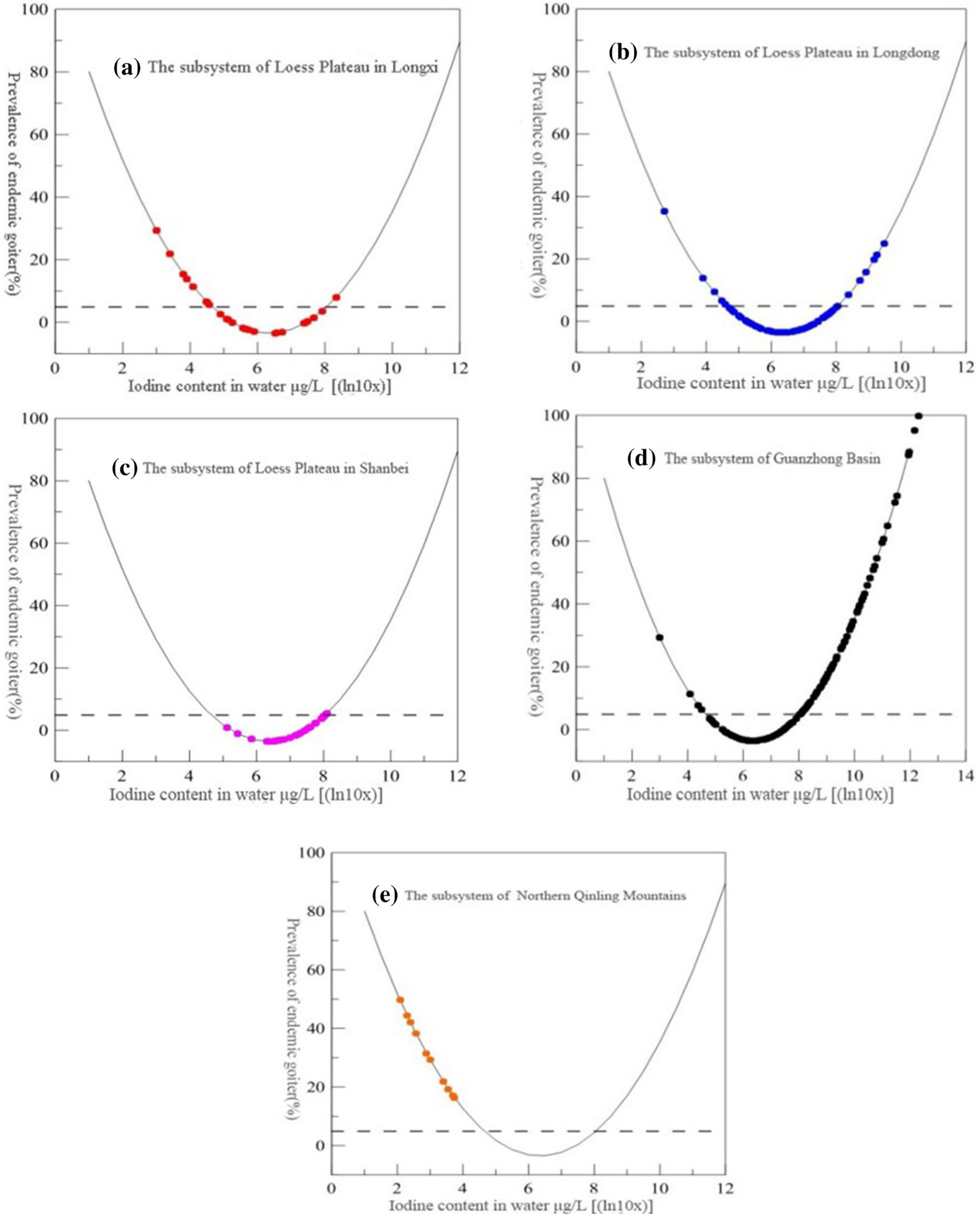

Fig. 6 Relationship between the disease prevalence and iodine content of groundwater in Wei River Basin

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