

Distribution and potential health risk of nitrate in centralized groundwater sources of Wanbei Plain, Central China

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ABSTRACT

Centralized groundwater sources are the main source for drinking, domestic, industry, and agriculture in Central China. Thirty-five centralized groundwater sources were selected to determine the concentration and distribution characterization of nitrate in Wanbei Plain, Anhui Province. Meanwhile, the health risk assessment model recommended by USEPA was used to evaluate the potential health risk for adults and children. The results showed that the average concentration of nitrate in the pore water is lower than that of karst water, which may be attributed to the thinner thickness of loose sediments for karst water. The nitrate in groundwater mainly originates from chemical fertilizers, industrial and domestic waste. The health risk of nitrate to children through drinking water intake and skin contact was significantly higher than that of adults, which was explained by their being more sensitive receptors and facing higher health risks than adults. The non-carcinogenic risk of nitrate in all samples was within the acceptable range. The results also indicated drinking water control is the main way to reduce nitrate health risk. Nevertheless, there is an urgent need to take measures to strengthen groundwater management, improve groundwater quality, and reduce nitrate health risk.

Key words | Central China, centralized groundwater, distribution, health risk, nitrate

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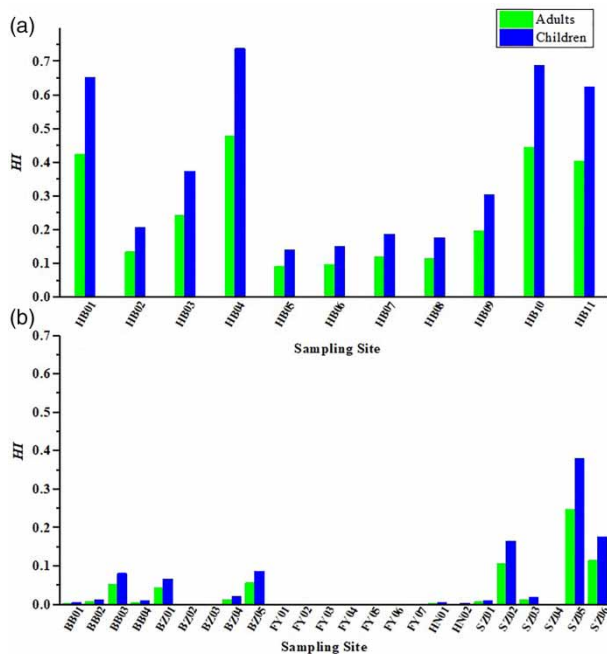
HIGHLIGHTS

- The average concentration of nitrate in the pore groundwater is lower than that of karst.
- Children are more sensitive receptors and face higher health risks than adults.
- Drinking water control is the main way to reduce nitrate health risk.
- All risks are within the acceptable range.
- Protective measures should be taken to avoid the nitrate pollution in groundwater and prevent its concentration from increasing.

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doi: 10.2166/aqua.2021.156

GRAPHICAL ABSTRACT



INTRODUCTION

Groundwater is the most abundant and widely distributed freshwater resource in the world (He *et al.* 2019; Das *et al.* 2020), and is of great significance for domestic and irrigation purposes (Teng *et al.* 2019). The enhancement of human activities has continued to exacerbate groundwater quality security problems (Ahada & Suthar 2018). With the rapid development of industry, agriculture and the acceleration of urbanization (Soldatova *et al.* 2017), the concentration of nitrate in groundwater is increasing gradually (Jin *et al.* 2015). Nitrate is the key pollutant in groundwater (Zhang *et al.* 2015), which has received worldwide attention (Goni *et al.* 2019). Groundwater nitrate sources mainly include sewage, livestock manure and toilet sewage, land degradation, atmospheric nitrogen deposition, and chemical fertilizer and organic fertilizer application (Chen *et al.* 2006; Deng *et al.* 2011; Czekaj *et al.* 2016; Zendeabad *et al.* 2019). The transformation process and the distribution characterization of nitrate in groundwater are mainly affected by physical processes (recharge rate, nitrogen application rate, mixing dilution, hydrological migration path)

and geochemical reaction processes (such as mineral composition and redox) (He *et al.* 2019). The nitrate pollution in groundwater is regionally different, which is mainly attributed to nitrate pollution sources, soil texture, climate conditions, groundwater depth, aquifer hydrogeological conditions, and redox reaction conditions during the process of nitrogen entering groundwater from the surface (Han *et al.* 2018; Kim *et al.* 2019).

Nitrate in groundwater can enter the human body through drinking water intake and skin contact, and has a direct impact on human health (Ma *et al.* 2016). According to previous studies (Rojas Fabro *et al.* 2015; Ahada & Suthar 2018; Qasemi *et al.* 2018), health impacts associated with the formation of strong carcinogen nitroso compounds include cancer, stomach cancer, colorectal cancer, lymphoma, malignant tumors, and blue baby syndrome (Gardner *et al.* 2020). Some studies show that the elevated incidence rate of cancer in many areas is closely related to the pollution of nitrate in drinking water (Chen *et al.* 2018; Qasemi *et al.* 2018). According to the regulations of the

World Health Organization (WHO), the maximum allowable concentration of nitrate nitrogen in drinking water is suggested as 10 mg/L in the United States and Japan, and 11.3 mg/L in European regulation (Sheng *et al.* 2019). The Ministry of Health P.R. China has set a guideline value of 20 mg/L for nitrate in public drinking groundwater sources (Luo *et al.* 2011). Owing to the serious effects of nitrate in groundwater on human health, it is of great importance to study the pollution of nitrate in drinking groundwater and health risk assessment.

The Wanbei Plain, located in Central China, is an important grain production base in China, with a large population density of 25,516 million. Groundwater is the main source for drinking, domestic, industry and agriculture in the area, and it plays an important role in social and economic development. The per capita water resource in Wanbei Plain is about 470 cubic meters, less than one-quarter of China's average level. At present, the water resources of Wanbei Plain are less than one-fifth of Anhui Province, but they support about half of the cultivated land and population in Anhui Province (Hu *et al.* 2015a). In recent years, with the increase of human activities, the nitrate content in groundwater has increased gradually and has seriously endangered the safety of drinking water (Hu *et al.* 2015b, 2017). Previous studies on groundwater in Wanbei Plain mainly focused on groundwater quality assessment, hydraulic connection, and distribution characteristics of the main indexes affecting groundwater quality (Wang *et al.* 2010, 2013; Yang *et al.* 2014). However, the problem of nitrate pollution in groundwater resources has not been paid enough attention, and there are few studies on nitrate health risk assessment in groundwater.

Health risk assessment is a successful method for assessing nitrate pollution and human health (Shalyari *et al.* 2019). It can quantitatively assess the probability of nitrate pollution in the human body and provide evidence for the relationship between nitrate pollution and incidence rate of cancer. It can provide important information to prevent and control nitrate pollution in groundwater and ensure drinking water security. In view of the different aquifer types in the study area, this study aimed to investigate nitrate contents in groundwater and to evaluate the health risk from drinking water and skin contact with contaminated water in Wanbei Plain. The objective was to compare and analyze

the health risk level of different exposure routes between adults and children. The results are expected to indicate specific regions which would help authorities, decision-makers, practitioners, and other professionals to take effective measures for the management of water supply projects and develop regional drinking water security planning.

MATERIALS AND METHODS

Study area

Wanbei Plain is located to the north of Huaihe River in Central China. It is adjacent to Jiangsu Province in the east, Henan Province in the west, and Shandong Province in the north. Its geographical coordinates are 114°50'–118°15'E and 32°25'–34°40'N, with a total area of 3.84×10^4 km². It belongs to the semi-humid monsoon climate zone of warm temperate zone, with annual average temperature, annual precipitation and evaporation of 14–15.5 °C, 800–1,000 mm, and 1,500–1,600 mm, respectively.

The groundwater bearing rock groups in the Wanbei Plain are mainly composed of karst water in carbonate aquifers and pore water distributed in loose Quaternary sediments. According to the underground conditions, the pore water bearing formations of loose Quaternary sediments are divided into shallow aquifer group and deep aquifer group. The groundwater is mainly recharged by shallow water overflow and lateral runoff, and the discharge mode is mainly artificial exploitation and lateral runoff. The karst aquifers are mainly distributed in Huaibei City, and are the main water supply layers for production and living in Huaibei City. The supply sources include atmospheric precipitation, surface water infiltration, and overlying pore aquifer overflow. The drainage mode is mainly artificial mining. The northern part of Wanbei Plain is exposed to surface forming hills and foothills, and the rest of the area is concealed below loose sediments.

Sample collection and analysis

A total of 35 samples were taken from the centralized water sources to determine the concentration and health risk of nitrate (Figure 1). Among them, 11 karst groundwater

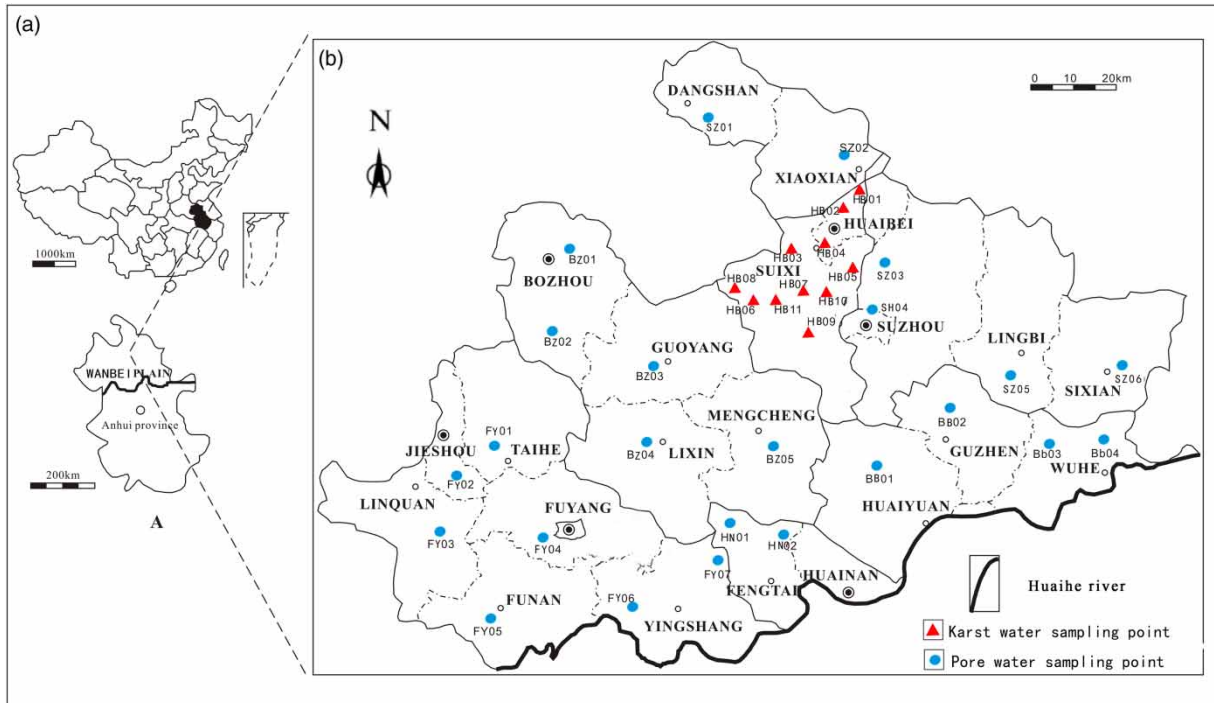


Figure 1 | (a) Location map and (b) sampling points of the study area.

samples from carbonate aquifers were collected in Huaibei City, and the other 24 samples were all taken from pore water distributed in loose Quaternary sediments. The sampling points were accurately located by GPS. All samples were collected in duplicate, one for analysis and the other for preservation for review. The sampling container was a polyethylene plastic bottle which was rinsed

with the groundwater for three to five times before sampling. All samples were sent back to the laboratory within 2 hours after adding concentrated sulfuric acid with $\text{pH} < 2$ and placed in the refrigerator (4°C) for testing. The water temperature, pH, conductivity, and dissolved oxygen were determined by using a multi-parameter meter (WTW3430, Germany) *in situ* (Figure 2). Nitrate concentrations of the



Figure 2 | Sample collection *in situ*.

samples were analyzed using UV-visible spectrophotometer (HACH DR/6000) with a detection limit of 0.20 mg/L. The collection, storage, and analysis methods of water samples are in accordance with the drinking water inspection standards set by the Ministry of Health, China in 2006.

Health risk assessment model

According to the harmful effects of different pollutants on the human body, various risk assessment models were established recently (Rojas Fabro *et al.* 2015). Among them, the internationally recognized health risk assessment model recommended by the United States Environmental Protection Agency (USEPA) has been widely used in the water quality assessment of drinking water (Shalyari *et al.* 2019). As it has been widely used in the evaluation of drinking water quality, this study used this model to evaluate the health risk of nitrate pollution in the groundwater of Wanbei Plain.

It is generally believed that somatic toxicity, the risk assessment of substances, is based on the reference dose (Teng *et al.* 2019). Toxic effects may occur when the exposure dose exceeds the reference dose. The evaluation model is

$$HI = \frac{I_{CD}}{D_{RF}} \quad (1)$$

where HI is the non-carcinogenic risk index (unitless); I_{CD} is the daily average exposure dose ($\text{mg}\cdot\text{kg}^{-1}\cdot\text{D}^{-1}$); and D_{RF} is the reference dose of a specific non-carcinogen in groundwater ($\text{mg}\cdot\text{kg}^{-1}\cdot\text{D}^{-1}$). The non-carcinogenic risk threshold recommended by USEPA is 1 (Suvarna *et al.* 2020). When $HI < 1$, the non-carcinogenic health risk caused by pollutants is within the acceptable range, and the health of the exposed person is not likely to suffer obvious adverse effects; the non-carcinogenic health risk caused by pollutants is unacceptable if $HI > 1$. The non-carcinogenic health risk tends to increase with the increasing of HI .

Nitrate in drinking water enters the human body mainly through drinking water intake and skin contact (Zhai *et al.* 2017). The calculation formula of daily average exposure

dose I_{CD} is defined as:

$$I_{CD} = I_{CDI} + I_{CDD} \quad (2)$$

$$I_{CDI} = \frac{C \cdot IR \cdot ABS \cdot EF \cdot ED}{BW \cdot AT} \quad (3)$$

$$I_{CDD} = \frac{C \cdot SA \cdot EV \cdot K_P \cdot ET \cdot EF \cdot ED \cdot CF}{BW \cdot AT} \quad (4)$$

where I_{CDI} is the daily average exposure dose of drinking water ($\text{mg}\cdot\text{kg}^{-1}\cdot\text{D}^{-1}$); I_{CDD} is the daily average exposure dose through skin contact ($\text{mg}\cdot\text{kg}^{-1}\cdot\text{D}^{-1}$); C is the measured concentration of nitrate in groundwater ($\text{mg}\cdot\text{L}^{-1}$); IR is the drinking water rate ($\text{L}\cdot\text{D}^{-1}$); ABS is the gastrointestinal absorption coefficient, which is related to pollutants; EF is the exposure frequency (assumed to be 365 days a year); ED is the exposure duration; BW is the average weight of residents (kg); SA is the skin contact surface area (cm^2); K_P is the skin permeability coefficient of pollutants ($\text{cm}\cdot\text{h}^{-1}$); EV is the frequency of bathing; ET is the bath time ($\text{h}\cdot\text{D}^{-1}$); AT is the time parameter of average exposure dose (d); CF is the volume conversion factor ($\text{L}\cdot\text{cm}^{-2}$). The calculation formula using the parameters were primarily selected from the USEPA guideline for risk assessment (Luo *et al.* 2011) and are listed in Table 1.

Table 1 | Parameters employed for human health risk assessment

Parameters	Children	Adults
DRf	1.6	1.6
C	Actual measurement	Actual measurement
IR	1.8	2
BW	35	60
ET	0.167	0.167
ABS	0.5	0.5
AT	365 × ED	365 × ED
EV	1	1
KP	0.001	0.001
CF	0.001	0.001
ED	30	30
EF	365	365
SA	10,000	16,500

RESULTS AND DISCUSSION

The concentration and distribution characterization of nitrate in groundwater

The concentration of nitrate in the karst water ranged from 8.80 to 45.89 mg/L, with the mean concentration of 24.01 mg/L (Table 2). In this research, the nitrate concentration of 10 pore groundwater samples in the loose layer

Table 2 | Nitrate content in groundwater

Groundwater type	Number of samples	Minimum	Maximum	Average
Karst water	11	8.8	45.89	24.01
Pore groundwater	24	–	23.63	4.62

–, below detection limit.

was lower than the detection limit, and the concentration of nitrate ranged from 0 to 23.63 mg/L with the mean concentration of 4.62 mg/L. The average concentration of nitrate in the pore water is lower than that of karst water. As shown in Figure 3, 9 of 11 karst water samples exceeded the Chinese drinking groundwater standard of 20 mg/L, accounting for 81.8%. Three of 24 pore groundwater samples exceeded the Chinese drinking groundwater standard, accounting for 12.5%.

The Wanbei Plain is an important agricultural base. A large number of chemical fertilizers and pesticides containing nitrogen are used and lead to the deposition of the nitrogen element, and eventually into the groundwater with precipitation, finally resulting in groundwater nitrate pollution. Meanwhile, livestock breeding is also an important source of nitrate in groundwater. Due to the backwardness of animal husbandry and breeding

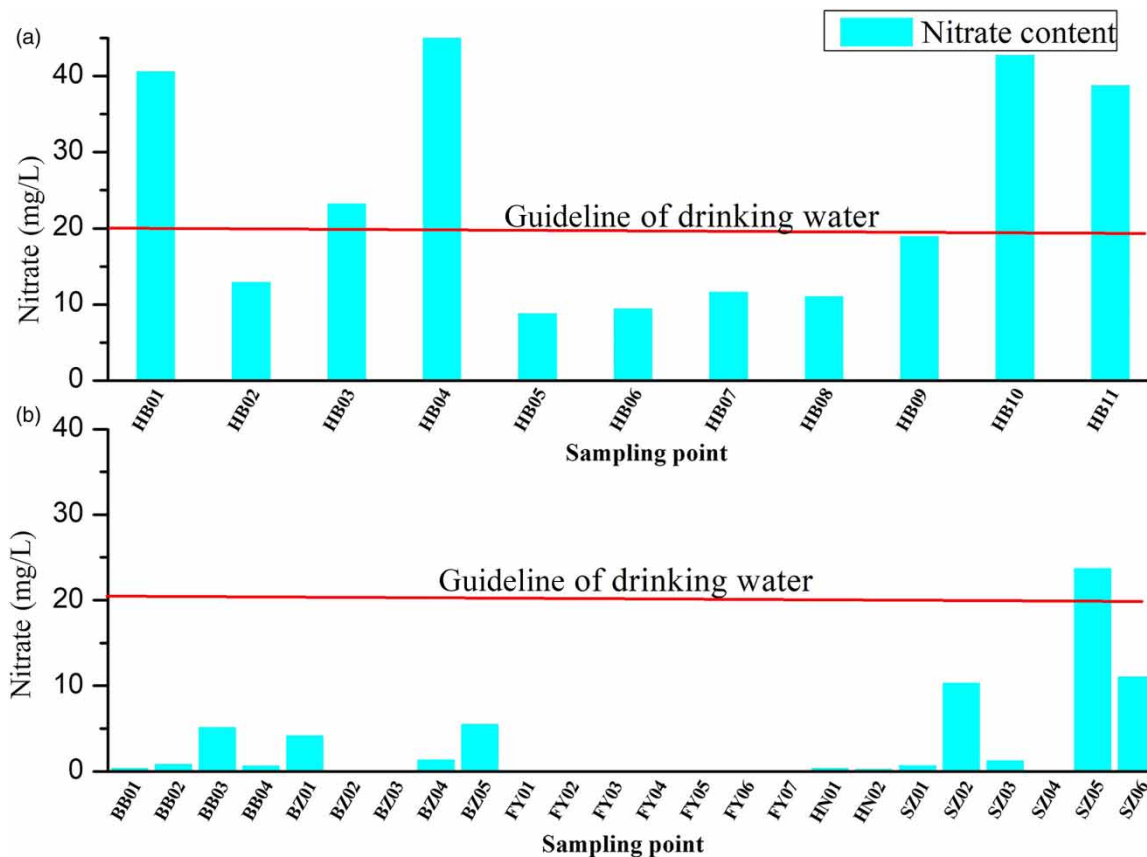


Figure 3 | Comparison of mean nitrate concentrations with guideline of drinking water with nitrate in centralized groundwater sources of Wanbei Plain: (a) karst groundwater and (b) pore groundwater.

technology in some rural areas, untreated poultry manure is randomly piled up. The nitrogen element in feces can directly enter into the soil for infiltration through rain washing and soil capillary action, causing the increasing of nitrate in groundwater (Wang *et al.* 2013).

Additionally, most of the nitrate in groundwater in Wanbei Plain originates from industrial and domestic waste. During the process of infiltration from the surface, nitrate will be subject to the nitrification and denitrification of soil nitrifying bacteria and denitrifying bacteria, and the adsorption of soil particles (Zheng *et al.* 2020). The concentration of nitrate decreases with the increasing of well depth (Zhang *et al.* 2017). The elevated concentration of nitrate in the selected karst groundwater may be explained by the thinner thickness of loose sediments in Huaibei City, nitrate nitrogen is not absorbed and enters directly into the karst aquifer, resulting in high nitrate content in groundwater (Hu *et al.* 2015a).

The potential health risk of nitrate in groundwater

Non-carcinogenic risk from drinking water

Figure 4 illustrates the values of *HI* comparison of mean nitrate concentrations in karst water and pore water samples. The results indicated that the risk of adverse health effects of nitrate in groundwater is within the acceptable range. The mean values of *HI* from nitrate in karst water for adults and children were 0.25 (0.09–0.48) and 0.39 (0.14–0.74), respectively. For nitrate in pore groundwater, the mean values of *HI* for adults and children were 0.03 (0–0.25) and 0.04 (0–0.38), respectively. The average health risk index of children who ingest nitrate through drinking water is greater than that of adults, which suggested that children are more sensitive receptors and face higher health risks than adults. Because the nitrate concentration in karst water is higher than that in pore water,

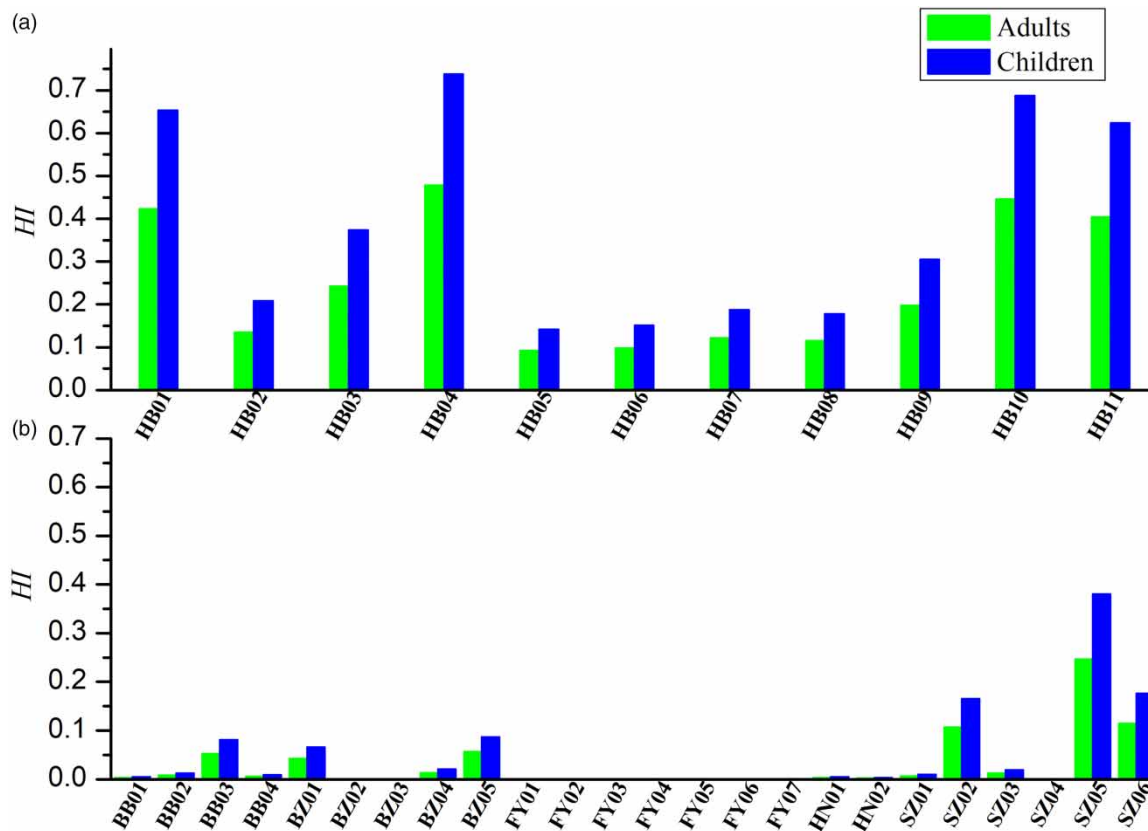


Figure 4 | Statistical graph of non-carcinogenic risk from drinking water of drinking water with nitrate in centralized groundwater sources of Wanbei Plain: (a) karst groundwater and (b) pore groundwater.

the former health risk value of individuals through drinking water is also greater than that of the latter. The health risk index of adults and children is less than 1, which indicates that the non-carcinogenic health risk caused by nitrate is within the acceptable range, and the health of contacts is unlikely to suffer obvious adverse effects. However, the groundwater nitrate concentration may be increasing quickly for an agricultural activity or unreasonable exploitation of groundwater, thus the groundwater nitrate pollution should be addressed carefully.

Non-carcinogenic risk of skin acceptance pathway

The values of *HI* by skin acceptance pathway were calculated separately for adults and children and are depicted in Figure 5. The mean values of *HI* from nitrate in karst water for adults and children were 6.89×10^{-4} (2.52×10^{-4} – 1.32×10^{-3}) and 7.16×10^{-4} (2.62×10^{-4} – 1.37×10^{-3}), respectively. For nitrate in pore groundwater, the mean values of *HI* for adults and children were 7.73×10^{-5} (0 – 6.78×10^{-4}) and 8.03×10^{-5} (0 – 7.05×10^{-4}), respectively. The non-carcinogenic risk of total health risk skin contact route is far less than 1, which indicates that the health risk of nitrates through skin contact is very small and could be regarded as negligible.

When comparing the non-carcinogenic risks of drinking water and skin contact, the health risks of children through drinking water intake and skin contact were significantly higher than those of adults, which may be related to the fact that the environmental pollution per unit weight of children is more sensitive than that of adults (Xu & Zhang 2018). Therefore, sufficient attention should be paid to the health risk of nitrate in groundwater to children.

Total non-carcinogenic risk

The total non-carcinogenic risk was calculated and presented in Figure 6. The mean values of *HI* from nitrate in karst water for adults and children were 2.51×10^{-1} (9.19×10^{-2} – 4.79×10^{-1}) and 3.87×10^{-1} (1.42×10^{-1} – 7.39×10^{-1}), respectively. For nitrate of pore groundwater, the mean values of *HI* for adults and children were 2.81×10^{-2} (0 – 2.47×10^{-1}) and 4.34×10^{-2} (0 – 3.81×10^{-1}), respectively. The total non-carcinogenic risk of karst water is higher than that of pore groundwater, and the total non-carcinogenic

risk to children is greater than that of adults. All the total non-carcinogenic risks for the selected samples are less than 1 and lower than the threshold value of human health hazard effect. The health risk values of some karst water samples are close to the non-carcinogenic risk threshold of 1, so measures should be taken to avoid the increase of nitrate concentration in groundwater.

It can be seen that the total risk of nitrate intake in the two exposure routes is roughly equal to the health risk of drinking water. The contribution rate of non-carcinogenic risk caused by drinking water accounted for 99.81%, suggesting that nitrate in groundwater mainly enters the human body through drinking water. The result is consistent with the results of other researchers (Zhai *et al.* 2017; Shalyari *et al.* 2019; Sheng *et al.* 2019). Therefore, drinking water control should be the main way to reduce the health risk of nitrate. Wanbei Plain is a base of agricultural production. The structure of agricultural production will not change greatly for a long time. The groundwater nitrate pollution caused by human activities, especially agricultural production, may exist for a long time. Therefore, it is still necessary to strengthen the prevention and control of groundwater pollution and risk management, to ensure the quality and safety of drinking water.

CONCLUSIONS

The average concentration of nitrate in the pore water is lower than that of karst water, which is mainly attributed to geological differences. The thickness of the unconsolidated layer of karst water in Huaibei is thinner than that of pore water layer areas. Nitrate could directly enter the karst aquifer without being adsorbed during nitrate infiltration, and thus resulting in high nitrate content in karst groundwater. The mean values of the total non-carcinogenic risk from nitrate in karst water for adults and children were higher than that of pore groundwater. The total non-carcinogenic risks of the selected samples were lower than the threshold value of human health hazard effect. The health risk of nitrate to children through drinking water intake and skin contact was significantly higher than that to adults. The contribution rate of non-carcinogenic risk caused by drinking water accounted for 99.81% of the

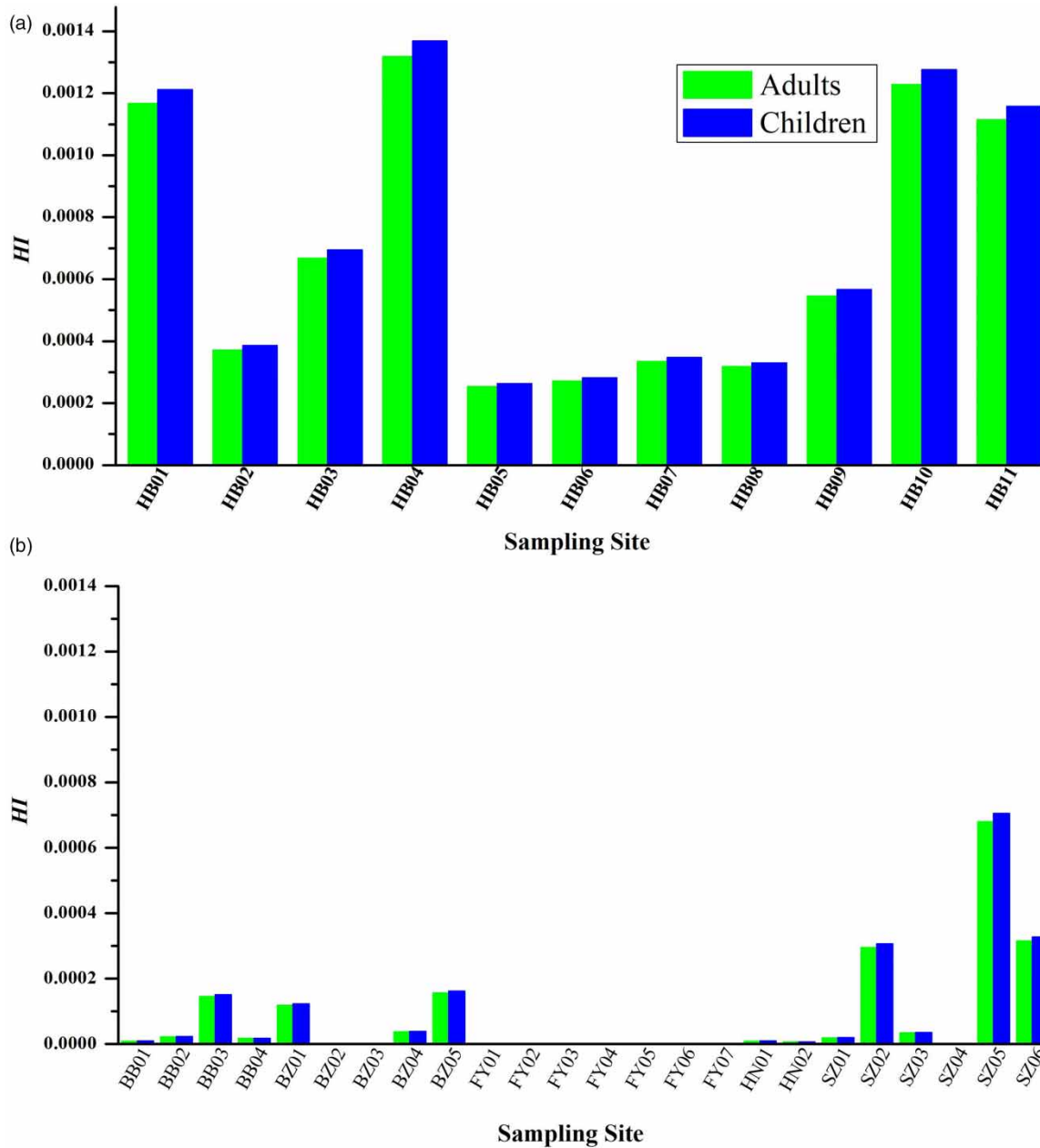


Figure 5 | Statistical graph of non-carcinogenic risk of skin acceptance pathway of water with nitrate in centralized groundwater sources of Wanbei Plain: (a) karst groundwater and (b) pore groundwater.

total risk, which was regarded as the main risk source, hence, drinking water control is the main way to reduce the health risk of nitrate. Therefore, stringent countermeasures should be adopted to ensure drinking water security.

This study evaluated the health risk caused by nitrate pollution, and provided the basis for the prevention and

control of nitrate pollution and health risk management. It is not only of great significance for the protection of groundwater safety and residents' health in this area, but also has a certain reference value for the development of groundwater nitrate pollution research and protection measures in other areas. In order to ensure the health and safety of drinking

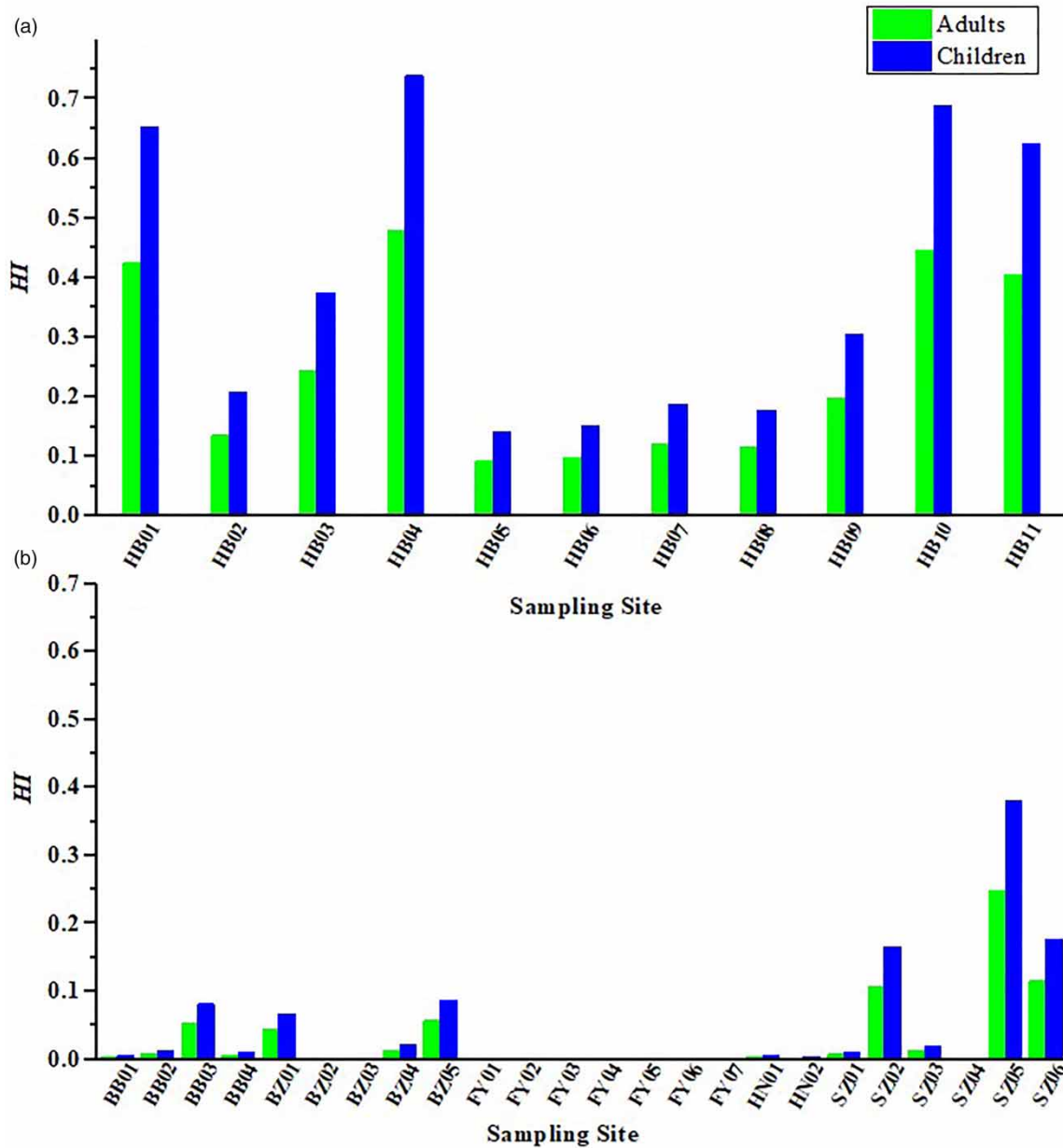


Figure 6 | Statistical graph of total non-carcinogenic risk of drinking water with nitrate in centralized groundwater sources of Wanbei Plain: (a) karst groundwater and (b) pore groundwater.

water, it is suggested that for the areas with zero health risk, protective measures should be taken to avoid the nitrate pollution in groundwater caused by human activities and prevent its concentration from increasing; for the areas with health risk, control or restoration measures should be taken to prevent the increase of health risk, and reduce the nitrate nitrogen concentration in groundwater and

reduce the pollution, slowing down the current state of health risk level.

Therefore, this evaluation is only a preliminary attempt, as there are many uncertain factors in health risk assessment. Drinking water health risk is closely related to people's consumption habits, occupation types, gender differences, and other factors. It is suggested to carry out

basic research on health risk assessment to provide suitable health risk for the population. and provide a scientific basis for safety management of regional groundwater quality.

ACKNOWLEDGEMENTS

This work was financially supported by the Key Program for Science and Technology Development of Anhui Province (No. 1908085MD113), the Postdoctoral Science Foundation of China (No. 2016M602037). We acknowledge editors and reviewers for polishing the language of the paper and for in-depth discussion.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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First received 24 November 2020; accepted in revised form 3 April 2021. Available online 30 April 2021