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Health risk analysis of nitrate in groundwater in Shanxi Province, China: A case study of the Datong Basin

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ABSTRACT

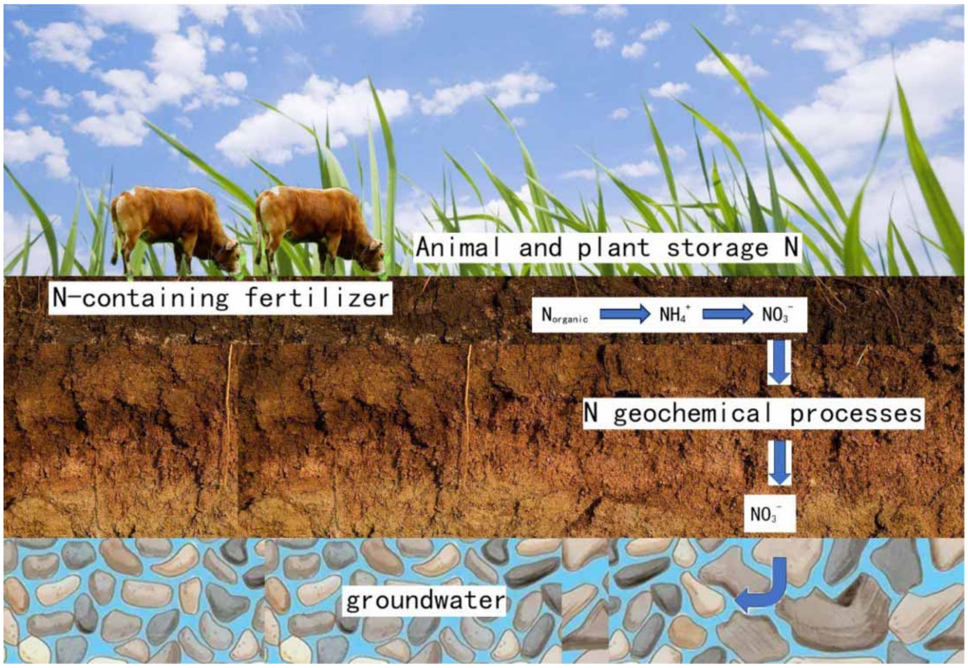
In order to identify and effectively control the impact of NO_3^- pollution on human health, on the basis of investigation, sampling, analysis and testing, statistical analysis software (SPSS19), groundwater pollution analysis software, Nemera comprehensive index method, correlation analysis method and human health risk assessment model are applied for analysis and research. The results indicate that the groundwater in the study area is mainly Class II water, with overall good water quality. The main influencing factors for producing Class IV are NO_3^-, Fe, F^- and SO_4^{2-}. The use of agricultural fertilizers is the main source of NO_3^- exceeding standards in groundwater in this area. There are significant differences in the health hazards caused by NO_3^- pollution in groundwater among different populations, and infants and young children are more susceptible to nitrate pollution. The division of pollution areas and high-risk groups plays an important guiding role in preventing health risks. The new achievements will help people improve their awareness of risk prevention, caring for the environment, respecting nature and implementing precise policies, promoting society to step onto the track of scientific and healthy development.

Key words: China, fertilizer overuse, HHRA, nitrate pollution, shallow groundwater

HIGHLIGHTS

- Scientific evaluation of groundwater quality was conducted and the main factors affecting water quality were analyzed.
- Scientific evaluation was conducted on NO_3^- pollution in the research area and the main sources of NO_3^- were identified.
- The significant differences in the health hazards of NO_3^- pollution were identified.
- The research results have an important guiding role in preventing health risks.

GRAPHICAL ABSTRACT



INTRODUCTION

Groundwater is one of the precious natural resources and the amount of groundwater resources accounts for one-third of the total water resources. Groundwater is closely related to people's production and life (Alireza *et al.* 2020; Tian *et al.* 2020a, 2020b, 2020c, 2020d), Groundwater is particularly important in areas with insufficient surface water resources (Tian *et al.* 2021). However, as the drinking water source of 70% of China's population, the groundwater has been polluted (Sun *et al.* 2020a, 2020b; Cao *et al.* 2023a), and the shallow groundwater and deep groundwater polluted by nitrides, phosphates and heavy metal ions have reached 80 and 30%, respectively (Sun *et al.* 2021; Cao *et al.* 2023b). Because groundwater pollution is highly hidden and difficult to detect, it is difficult to control the pollution in groundwater (Cao *et al.* 2018; Saravanan *et al.* 2022).

Nitrate pollution in groundwater is a worldwide environmental problem. Nitrate is easy to dissolve and has strong fluidity (Gu *et al.* 2013; Zhai *et al.* 2015; Tian *et al.* 2020a, 2020b, 2020c, 2020d). It can enter underground through leaching and surface runoff (Li *et al.* 2015). It is a chemical pollutant widely existing in groundwater (Hand *et al.* 2013; Sun *et al.* 2019). Drinking groundwater polluted by nitrate for a long time will lead to digestive system cancer, methemoglobinemia, blue baby syndrome and other diseases posing a serious threat to human health (Han 2014; China 2016; Sun *et al.* 2017; Chen *et al.* 2020). Nitrate-contaminated groundwater will cause harm when it enters the human body, therefore, it is necessary to evaluate the harm and degree of nitrate-contaminated groundwater. Human Health Risk Assessment (HHRA) is a qualitative and quantitative assessment method for the hazard degree of polluted groundwater to human body (Ni *et al.* 2010; Sun *et al.* 2022a, 2022b).

In 1983, the National Academy of Sciences (NAS) proposed for the first time a 'four-step method' based on hazard identification, dose–effect assessment, exposure assessment and risk characterization to assess the risks of environmental pollutants to human health. Then the United States Environmental Protection Agency (USEPA) made a more detailed description of health risk assessment. At present, the 'four-step' health risk assessment model is applied to the health risk assessment of various pollutants in various environmental media. Saravanan *et al.* (2022) and Zhang *et al.* (2021) used models to evaluate the impact of high nitrate in groundwater on human health in different regions.

Algorithms and models have been widely applied in various fields such as resource allocation (Deng *et al.* 2022a; Xu *et al.* 2023), public management (Deng *et al.* 2022b, 2022c) and health services (Duan *et al.* 2022; Huang *et al.* 2023). The article selects the HHRA model recommended by the USEPA to identify and assess the risk of nitrate pollution in groundwater in the Datong Basin. This helps to develop effective risk management measures, achieve sustainable and healthy development and achieve good results. Through model sensitivity analysis, the model is sensitive to the influence of pollutant concentration. Simulation methods can help us better understand the process of groundwater pollution and provide a basis for formulating effective prevention and control measures. The risk assessment method for groundwater pollution can quantitatively evaluate the degree of groundwater pollution risk and provide scientific decision-making basis for decision-makers.

According to the research on the Datong Basin from 2020 to 2021, there are areas with high nitrate content in the groundwater in the Datong Basin. The maximum nitrate content (N) reaches 474 mg/L. The high concentration of nitrate groundwater will affect human health after long-term drinking. In order to improve people's risk awareness, scientifically and reasonably prevent the occurrence of risks and ensure that human health is not harmed, it is necessary to conduct a health risk assessment of groundwater in the Datong Basin.

Through research, the health risk model was first introduced into the health risk assessment of groundwater pollution in the Datong area of Shanxi Province. The current water quality status, water quality impact factors, NO_3^- exceeding the standard status and health risks caused by NO_3^- exceeding the standard were identified in the study area. The conclusion that NO_3^- exceeding the standard has different health risks for different populations was drawn, which has practical significance for guiding people to scientifically and reasonably plant crops and use fertilizers in an appropriate amount.

The establishment of the model is based on three exposure environments: drinking water, skin and respiration. The model also has certain limitations. In the future, we should further research and develop simulation and evaluation methods to improve the accuracy and reliability of prediction and evaluation, in order to protect the sustainable use and management of groundwater resources.

MATERIALS AND METHODS

Study area

The Datong Basin is located in Shanxi Province, China, 112°15′–114°17′ E and 39°05′–40°33′ N. The total area of the plain area is about 7,440 km^2. The climate is characterized by dry climate, less rain, more sandstorms, large temperature difference between day and night, and sufficient sunlight. The average annual precipitation is between 363 and 414 mm. It belongs to the Haihe River system, the Sanggan River is the main river, and its tributaries include the Huihe River, the Huangshui River, the Shili River, the Yuhe River and the Hunhe River. The Datong Basin is surrounded by mountains, of which the highest peak is 2,426 m above sea level. The terrain in the center of the basin is open and flat, with an altitude of 1,100–950 m. At present, it has formed a relatively complete industrial system focusing on coal production, such as electric power, chemical industry, building materials, machinery and food. It is one of the energy and heavy chemical bases in China. The basin is densely populated. According to statistics, the current population of the Datong Basin is about 2.6 million. The Datong Basin is a Cenozoic graben type fault basin developed on the Mesozoic fold basement. The NE, EW and SN trending faults form the basin boundary, respectively. The groundwater aquifer system includes the karst aquifer system, the basalt fissure pore aquifer system, the bedrock fissure aquifer system and the quaternary pore aquifer system. Human activities have caused groundwater pollution, especially the excessive use of nitrogen fertilizer in agricultural production.

Sampling and measurements

Groundwater samples were distributed in the study area, mainly from rural self use wells, and some are centralized water supply wells. The sampling time was from October 1, 2020 to November 28, 2020. In total, 100 samples (as shown in Figure 1) were collected and sent for analysis according to the standard, which accurately reflected the current situation of local groundwater. Each sampling point pumped water for 10 min before sampling. Before sampling, the sample bottle was washed with well water for three times, then sampled and sealed. After taking the sample, the sample bottle was immediately stored in a 4 °C incubator for subsequent analysis.

The water sample test was completed by the testing agency with the national certification, and the sample preparation was carried out in strict accordance with the geological and mineral industry standard of the People's Republic of China, 'Method for Analysis of Groundwater Quality' DZ/T 0064. According to different test items, the water samples are subject to evaporation, color reagent addition, acidification, reduction and other treatments. Cl^-, SO_4^{2-}, F^- and NO_3^- were determined by ion chromatography (IC); Ca^{2+}, Mn and TFe were determined by inductively coupled plasma emission spectrometer (ICP-OES), and total dissolved solids (TDS) were determined by the gravimetric method. The total hardness was determined by EDTA-2Na titration. The pH value was measured by the glass electrode method.

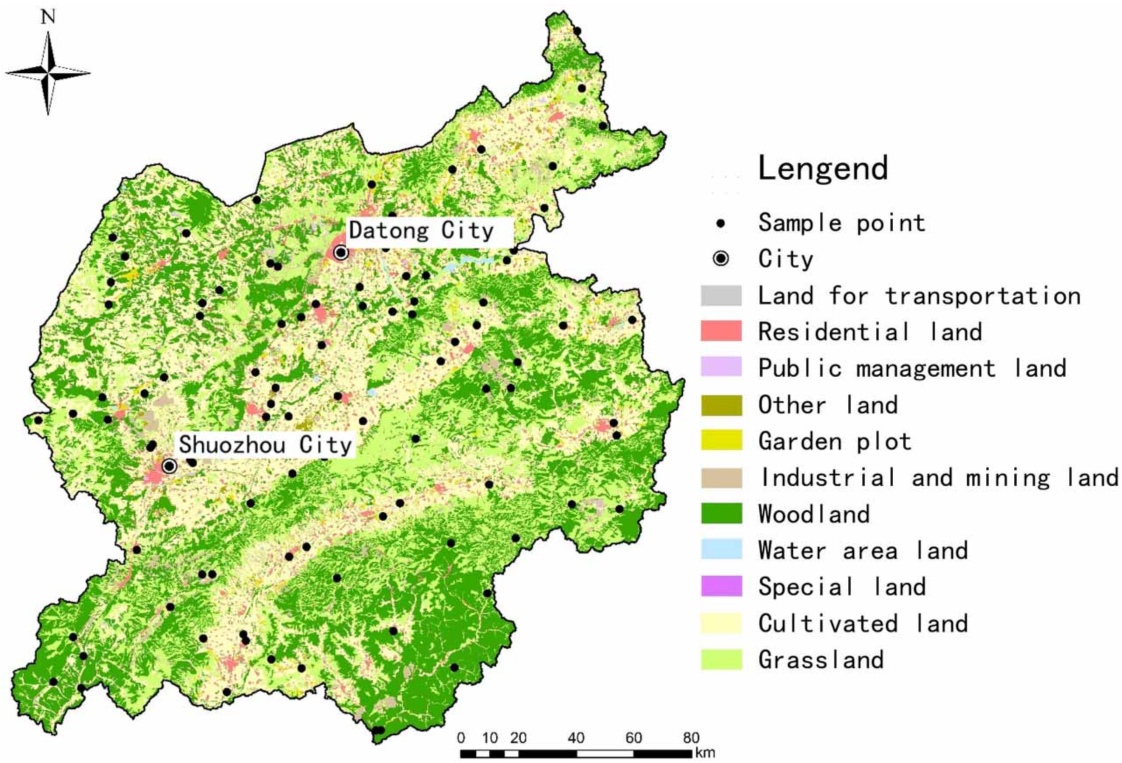


Figure 1 | Distribution of sampling points.

To ensure the accuracy and reliability of the analysis method, the spiked recovery method was used for determination. Each sample was analyzed 12 times, and the average analysis value and spiked recovery rate were calculated. The recovery rate (average of 12 times) of the item for spiked recovery determination should be controlled between 90 and 110%, and the relative standard deviation (RSD%) of 12 analyses of a single standard sample (or spiked recovery sample) should be calculated, with a requirement of RSD% $\leq$ 15%.

In total, four standard control samples and standard samples, such as GBW08606, were inserted into each batch of samples (control different items separately) and the accuracy of five recovered samples were analyzed and controlled simultaneously. The requirements of the specifications were strictly followed and the recovery rate of spiking was controlled at 80–120, 90–110 and 95–105%, respectively, according to different spiking quantities.

Standard sample monitoring and anion and cation balance monitoring are used for NO_3^- and other projects. The absolute value of the relative error E of the anion and cation balance was less than 5%, which is reliable data. After testing, all data obtained were reliable.

HHRA MODEL AND PARAMETER ACQUISITION

Groundwater quality health risk assessment is an effective method to quantitatively evaluate the hazard degree of hazardous substances in groundwater to human body. Generally, the assessment starts from hazard identification and then carries out dose–effect analysis to evaluate exposure and risk characterization (Zhou *et al.* 2016; Tian *et al.* 2020a, 2020b, 2020c, 2020d). On the basis of field investigation, sampling and indoor analysis (hazard determination), combined with the information on the harmful effects of chemicals on human health provided by the International Center for Research on Cancer (IARC), determine the health effects of nitrate on humans (dose–effect relationship) (Chen 2010; Chen *et al.* 2016).

At present, there are many risk models for evaluating the hazards of groundwater pollutants to human beings, among which the HHRA model recommended by the USEPA (Chen *et al.* 2017; Zhai *et al.* 2017; Javaid *et al.* 2019) is the most widely used in groundwater pollutant risk assessment (Su *et al.* 2013; Zhang *et al.* 2017; Mahmoud *et al.* 2018). In this paper, the health risk caused by nitrate pollution in groundwater in the Datong Basin is scientifically evaluated by using the evaluation model.

Hazard identification

The first step in HHRA is to identify hazards, that is, to determine the potential adverse effects of human intake of hazards, the possibility of such adverse effects, and the certainty and uncertainty of such adverse effects. The objective is to evaluate the evidence weight of adverse health effects according to the evaluation results of all existing toxicity and action mode data.

In order to achieve this goal, more detailed site-related data and historical information, concentration data of pollutants in groundwater and other samples of the site, analysis data of physical and chemical properties of the site, climate, hydrological and geological characteristics information and data of the site (location), relevant information such as land use mode, sensitive people and buildings of the site and surrounding plots are needed.

Dose–response assessment

The four-step model of population health risk assessment of the USEPA defines the dose–response assessment as 'describing the possibility and severity of adverse health effects under a certain exposure dose and exposure conditions of a chemical substance'. Dose–response relationship assessment provides a mathematical basis for converting exposure information to assess health risk. The dose–response relationship can be expressed by reference dose (RfD) (USEPA 2005). The determination of RfD is as follows:

$$\mathrm{RfD} = \frac{\mathrm{NOAEL(LOCAEL)}}{\mathrm{UFs}} \tag{1}$$

where RfD is the chronic reference dose (mg/kg d); LOCAEL is the lowest observed adverse effect level (mg/kg d); NOAEL is the no adverse effect level observed (mg/kg d) and UFs is the uncertainties.

The standard value of nitrate is 10 mg/L (USEPA 2001; Yang *et al.* 2012).

Exposure assessment

Exposure assessment is a process of measuring, estimating or predicting the intensity, time and frequency of people's exposure to pollutants in the medium. Exposure assessment is the quantitative basis for risk assessment, which is mainly the assessment of exposure environment, environmental concentration, receptor exposure route, environmental medium and exposure.

According to the definition of USEPA, skin absorption, air inhalation and direct drinking water intake were originally considered in the groundwater health risk assessment model. Since the nitrogen in groundwater does not volatilize, this paper only considers drinking water intake and skin intake, not air inhalation (Dzulfakar *et al.* 2011). Its calculation formula is:

$$I_{\mathrm{CDI}} = \frac{\mathrm{C} \times \mathrm{IR} \times \mathrm{ABS} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \tag{2}$$

$$I_{\mathrm{CDD}} = \frac{\mathrm{C} \times \mathrm{SA} \times K_p \times \mathrm{EV} \times \mathrm{ET} \times \mathrm{EF} \times \mathrm{ED} \times \mathrm{CF}}{\mathrm{BW} \times \mathrm{AT}} \tag{3}$$

where I_{CDI} is the daily average exposure dose through drinking water (mg/kg d); I_{CDD} is the daily average exposure dose of skin contact route (mg/kg d) and C is the measured concentration of nitrate in groundwater (mg/L).

The values of the parameters are shown in Table 1.

Risk characterization

Risk characterization is a process to estimate the probability of adverse health reactions of people exposed to target pollutants under various conditions. It is the last step of risk assessment. In this step, the data and analysis of the first three steps are integrated to estimate and predict the probability of response to the health effects of the exposed population caused by groundwater nitrate pollutants or the probability of the expected hazard level (Dzulfakar *et al.* 2011; Huang *et al.* 2018). It can be expressed as:

$$\mathrm{HI}_{\mathrm{oral-water}} = \frac{I_{\mathrm{CDI}}}{\mathrm{RfD}_{\mathrm{oral-water}}} \tag{4}$$

$$\mathrm{HI}_{\mathrm{derm-water}} = \frac{I_{\mathrm{CDD}}}{\mathrm{RfD}_{\mathrm{derm-water}}} \tag{5}$$

$$\mathrm{HI} = \mathrm{HI}_{\mathrm{oral-water}} + \mathrm{HI}_{\mathrm{derm-water}} \tag{6}$$

Table 1 | Meaning and value of each parameter

Parameter meaning		Value				Unit
		Children	Females	Males	Infants	
EF	Exposure frequency	365	365	365[b]	365	D/a
BW	Average body weight	32.02[e]	60.4[a]	69.55[a]	7.68[e]	kg
ABS	Gastrointestinal absorption coefficient	0.5[c]	0.5[c]	0.5[c]	0.5[c]	
IR	Amount of drinking water	1.5[b]	2[f]	2[f]	0.65[b]	L/d
ED	Exposure duration	6[b]	30[b]	30[b]	0.5[d]	a
SA	Body surface areas	9,035.2	1,600[a]	1,700[a]	3,416	cm^2
AT	Average exposure time	2,190	10,950	10,950	182.5[b]	d
EV	Bathing frequency	1[b]	1[b]	1[b]	1[b]	Time/d
ET	Bath time	0.167[c]	0.167[c]	0.167[c]	0.167[c]	h/d
CF	Unit conversion facter	0.002[b]	0.002[b]	0.002[b]	0.002[b]	L/cm^3
KP	Dermal adsorption	0.001[b]	0.001[b]	0.001[b]	0.001[b]	Cm/h

[a]These data come from PRCMEP (2014).
[b]These data come from USEPA (1989).
[c]These data come from USEPA (1991).
[d]These data come from USEPA (2004).
[e]These data come from PRCMH (2003).
[f]These data come from Tian *et al.* (2020a, 2020b, 2020c, 2020d).

where $HI_{oral-water}$ is the non-carcinogenic oral hazard coefficient (dimensionless); $HI_{derm-water}$ is the non-carcinogenic skin hazard coefficient (dimensionless); $RfD_{oral-water}$ is the reference dose of drinking water (mg/kg d), taken as 1.6; $RfD_{derm-water}$ is the reference dose absorbed by skin (mg/kg d), taken as 1.0; I_{CDI} is the intake dose of drinking water (mg/kg d); I_{CDD} is the skin absorbed dose (mg/kg d) and HI is the total risk coefficient (dimensionless).

For groundwater risk control value based on non-carcinogenic effect, the non-carcinogenic risk threshold recommended by USEPA is 1 (Huang *et al.* 2017; Javed *et al.* 2018; Wu *et al.* 2018).

In order to provide targeted and effective protection according to different groups of people, this paper carries out NO_3^- risk assessment for infants (one year old and below), children (2–17 years old), adult males and adult females, respectively. See Table 1 for specific parameters.

Data analysis

The distribution of groundwater chemical concentration and the correlation of its components were analyzed by statistical analysis software (SPSS 19). This method reveals the correlation between different chemical components of groundwater.

Water quality analysis

The quality standard for groundwater of the People's Republic of China (QSGC) is adopted for evaluation (GB/T14848-2017) (National Standard of the People's Republic of China 2017).

For the comprehensive evaluation of groundwater quality, the scoring method with notes is used to evaluate the individual components of the participating projects, divide the levels of the components and determine the evaluation score F_i of the individual components, respectively. According to the standards in Table 2, the formula for calculating the F value of

Table 2 | Quality evaluation and grading of single component

Category	I	II	III	IV	V
F_i	0	1	3	6	10

comprehensive evaluation score is as follows:

$$F = \sqrt{F_{avg}^2 + F_{max}^2} \tag{7}$$

where F is the comprehensive evaluation score; $F_{avg} = \sum F_i/N$ is the average value of evaluation score F_i of each single component; F_{max} is the maximum value in the evaluation score F_i of single component and N is the number of participating projects.

Then, according to the F value, the groundwater quality is classified according to the method specified in the standard (see Table 3).

RESULTS AND DISCUSSION

General characteristics of groundwater quality

The pH value is a comprehensive index to evaluate the acidity and alkalinity of aqueous solution. The maximum pH value of groundwater in the study area is 8.64, the minimum value is 7.42 and the average value is 8.13. On the whole, it is weakly alkaline and the coefficient of variation (CV) is small indicating that the pH value of groundwater does not change significantly. TDS refers to the total dissolved solids in the aqueous solution, $CaCO_3$ refers to the total hardness of the water, Fe refers to the total iron content in the study area, including F^-, SO_4^{2-} and Cl^- in the groundwater, which meet the requirements for drinking water and industrial water and meet the needs of human health. The average concentration of NO_3^- in the study area is 31.06 mg/L, which has exceeded the standard for healthy drinking water. The maximum concentration of NO_3^- is 474 mg/L, which is 15 times higher than the standard value and the CV value reaches 2 indicating that NO_3^- is distributed unevenly and has health risks (Table 4 and Figure 2).

Nitrate pollution of groundwater

Figures 3 and 4 show that NO_3^- Class I area accounts for 6% of the sample data, Class II area accounts for 8% of the sample data, Class III area accounts for 52% of the sample data, Class I–III water is mainly distributed in Shuozhou, Daixian and Wutai Area, Class IV area accounts for 13% of the sample data, mainly distributed in Datong, Hunyuan and Lingqiu, Class V area accounts for 21% of the sample data, mainly distributed in Zuoyun and Yingxian areas, as well as Yanggao and Guangling areas. The results show that the nitrate pollution degree of groundwater in this area is high and has zoning. Overall, the over standard rate of NO_3^- is 34%. The high content of NO_3^- should be highly valued and health risk assessment should be carried out.

Table 3 | Classification of comprehensive evaluation of groundwater quality

Code	I	II	III	IV	V
Level	Excellent	Good	Preferably	Poor	Range
Pollution degree	Uncontaminated	Slight pollution	Medium pollution	Heavy pollution	Severe pollution
F	<0.80	$\geq 0.80, <2.50$	$\geq 2.50, <4.25$	$\geq 4.25, <7.20$	≥ 7.20

Table 4 | Chemical characteristic values of groundwater in the study area (pH: dimensionless, other material units: mg/L)

	$CaCO_3$	TDS	pH	Cl^-	SO_4^{2-}	NO_3^-	Fe	F^-
MIN	129	198	7.42	4.17	10.8	0.11	0.019	0.34
MAX	1,175	1,935	8.64	302	1,074	474	0.64	2.25
AVG	295.61	512.82	8.13	39.4	105.53	31.06	0.1	0.89
SV	$\leq$450	$\leq$1,000	$6.5 \leq pH \leq 8.5$	$\leq$250	$\leq$250	$\leq$20.0	$\leq$0.3	$\leq$1.0
CV	0.55	0.68	0.02	1.55	1.3	2	1.18	0.44

Notes: MIN, minimum value; AVG, average value; MAX, maximum value; CV, coefficient of variation; SV, Standard value.

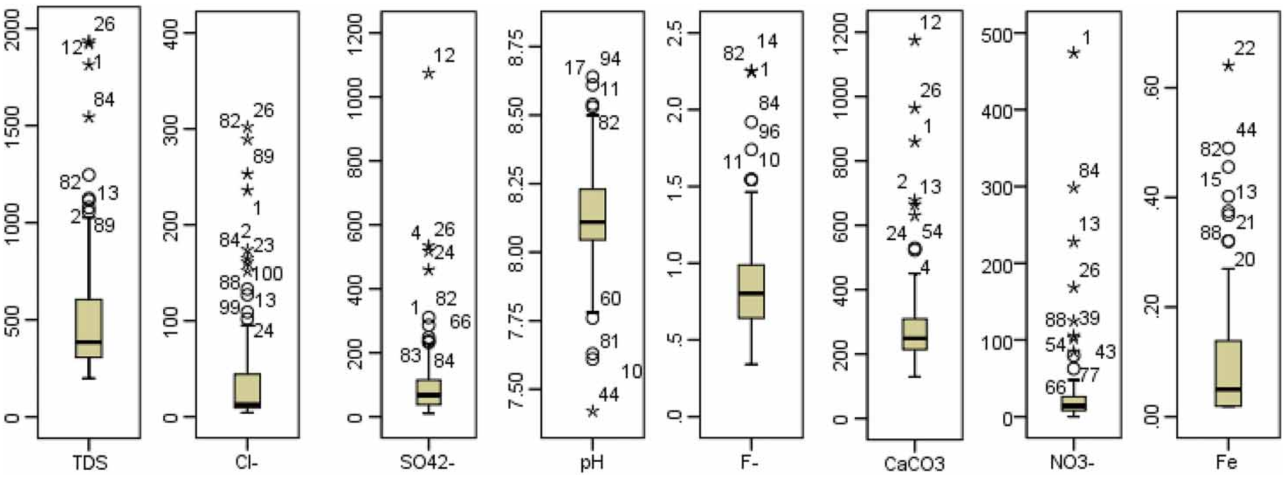


Figure 2 | Chemical concentration distribution map of groundwater (pH: dimensionless, other material units: mg/L).

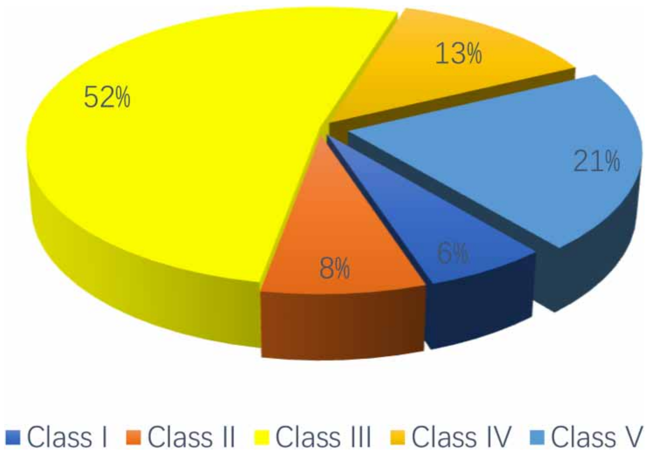


Figure 3 | Proportion of various types of nitrates.

Groundwater quality evaluation

With the development of industry and agriculture, the discharge of industrial wastewater and the continuous use of agricultural fertilizers and pesticides, the groundwater quality in the study area is deteriorating, and the groundwater pollution continues to occur and develop. Finding out the current situation and distribution of groundwater quality can effectively support the construction of national ecological civilization and provide basis for ecological restoration.

According to QSGC, the quality of groundwater samples is evaluated by using groundwater analysis software and the Nemero index method. The comprehensive evaluation results show that there are four levels of groundwater quality, Class I water quality accounts for 4%, Class II water quality accounts for 57%, Class III water quality accounts for 9%, Class IV water quality accounts for 30% and Class V water quality is not evaluated (Figure 5).

From the distribution area, there are Class I water in Jingping Town, Caicun Town, Xiliu village township and Xijiaoshan township, Class II water in Tianzhen County, Class III water in Hunyuan district and Zuoyun Xinrong district, Class IV water in Shentou spring area, the Datong Basin, Lingqiu district and Yuanping district. The groundwater in the study area is mainly Class II water, with good overall quality and no Class V water, however, Class IV water accounts for a large proportion, and monitoring and repair need to be strengthened (Figure 6). The main influencing factors of water quality are NO_3^-, Fe, F^-, SO_4^{2-}, total hardness ($CaCO_3$) and TDS. Through the research, the current situation and distribution characteristics of

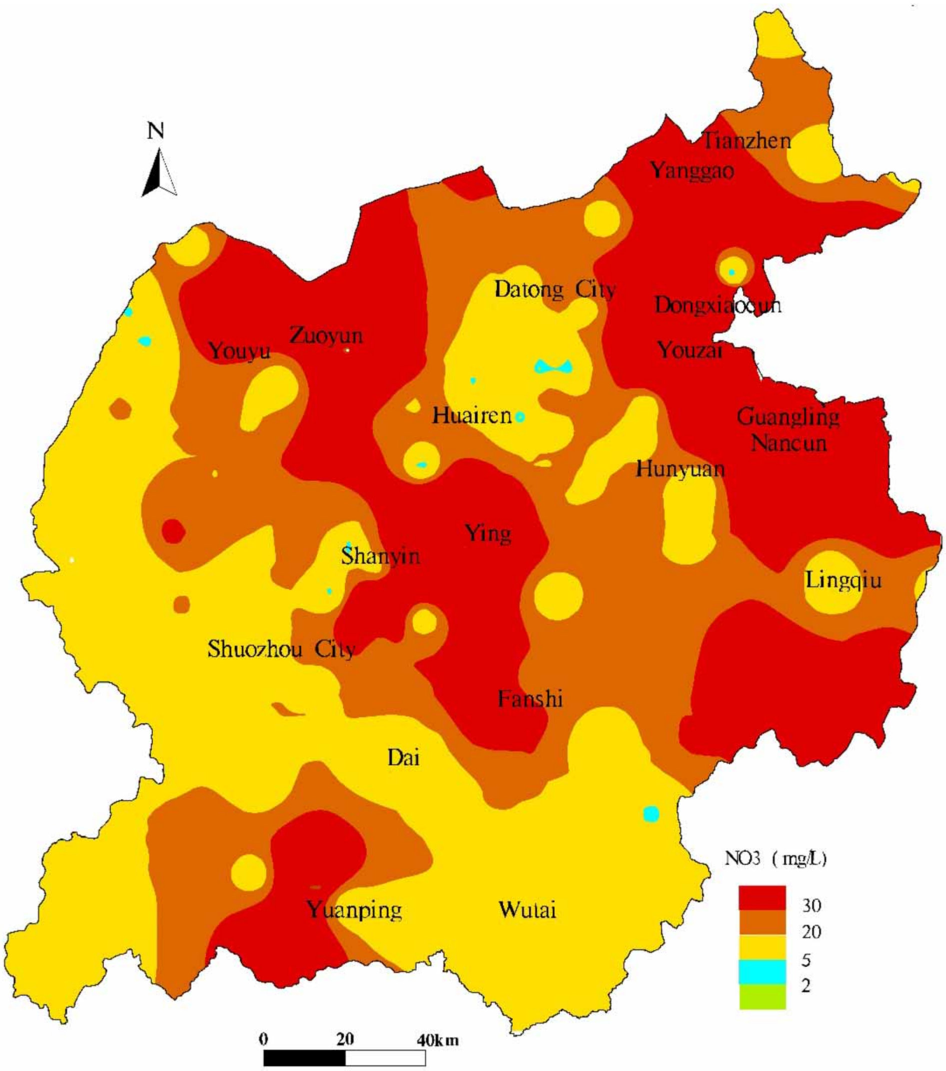


Figure 4 | Zoning map of nitrate pollution.

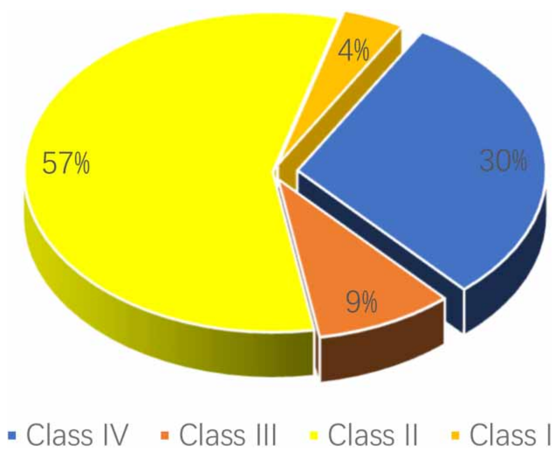


Figure 5 | Pie chart of groundwater quality type.

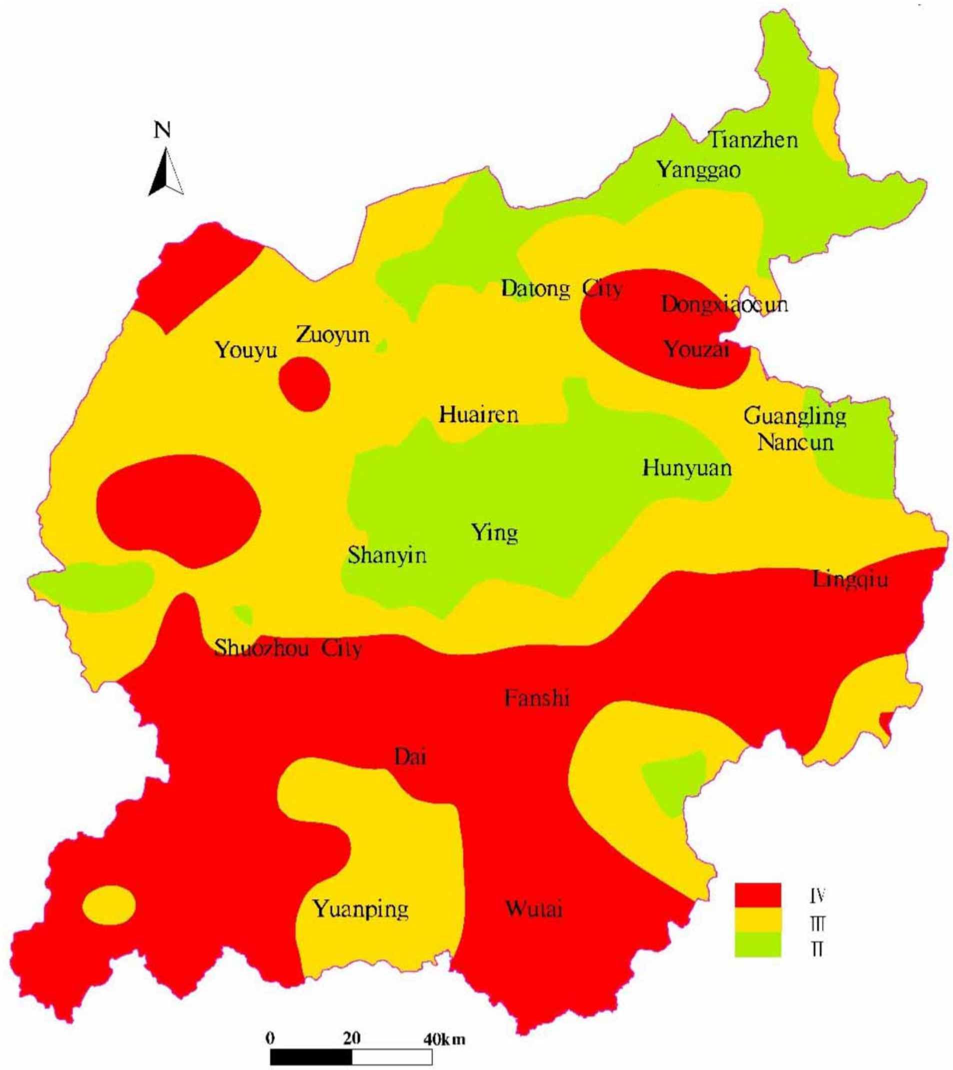


Figure 6 | Zoning of groundwater quality.

groundwater quality are found out, which can effectively support the construction of national ecological civilization and provide a reliable basis for ecological restoration.

SPSS19 software was used for correlation analysis of eight parameters: NO_3^-, TDS, Cl^-, $CaCO_3$, pH, F^-, Fe and SO_4^{2-} (Table 5) (Nakagawa *et al.* 2016). NO_3^- was significantly correlated with $CaCO_3$ and Cl^- at 0.01 level, and NO_3^- and SO_4^{2-} were significantly correlated at 0.05 level (Jalali 2011). The significant correlation between NO_3^- and $CaCO_3$, Cl^-, SO_4^{2-} indicates that the increase in NO_3^- concentration is related to the use of NO_3^- fertilizers and organic fertilizers in agricultural production, as well as the discharge of sewage in people's lives (Rahmati *et al.* 2014; Lasagna *et al.* 2016). Therefore, it is believed that the unreasonable use of fertilizers and the discharge of sewage in human production and life are the main reasons for the increase in NO_3^- concentration in groundwater (Li *et al.* 2016).

HEALTH RISKS FOR ADULT WOMEN AND MEN

HHRA health risk calculation results are shown (Figure 7). The maximum HI value of adult males is 8.55; there are health risks in 10% of the study area. In other words, NO_3^- pollution of groundwater usually does not cause harm to adult males in the area. However, in Yingxian County, Dongxiao village and Nancun village, NO_3^- pollution has health risks. The maximum HI value of adult women is 9.84 and about 10% of the study area has health risks. It is distributed in Yingxian County,

Table 5 | Correlation table of water chemistry parameters

	$CaCO_3$	TDS	pH	Cl^-	SO_4^{2-}	NO_3^-	Fe	F^-
$CaCO_3$	1							
TDS	.838**	1						
pH	−.200*	−0.04	1					
Cl^-	.474**	.782**	0.049	1				
SO_4^{2-}	.804**	.797**	−0.04	.433**	1			
NO_3^-	.518**	.656**	−0.06	.525**	.231*	1		
Fe	0.007	0.081	−0.11	0.146	−0.02	0.066	1	
F^-	0.064	.390**	.311**	.394**	0.126	.313**	0.15	1

*At 0.05 level, there was significant correlation.
**At 0.01 level, there was significant correlation.

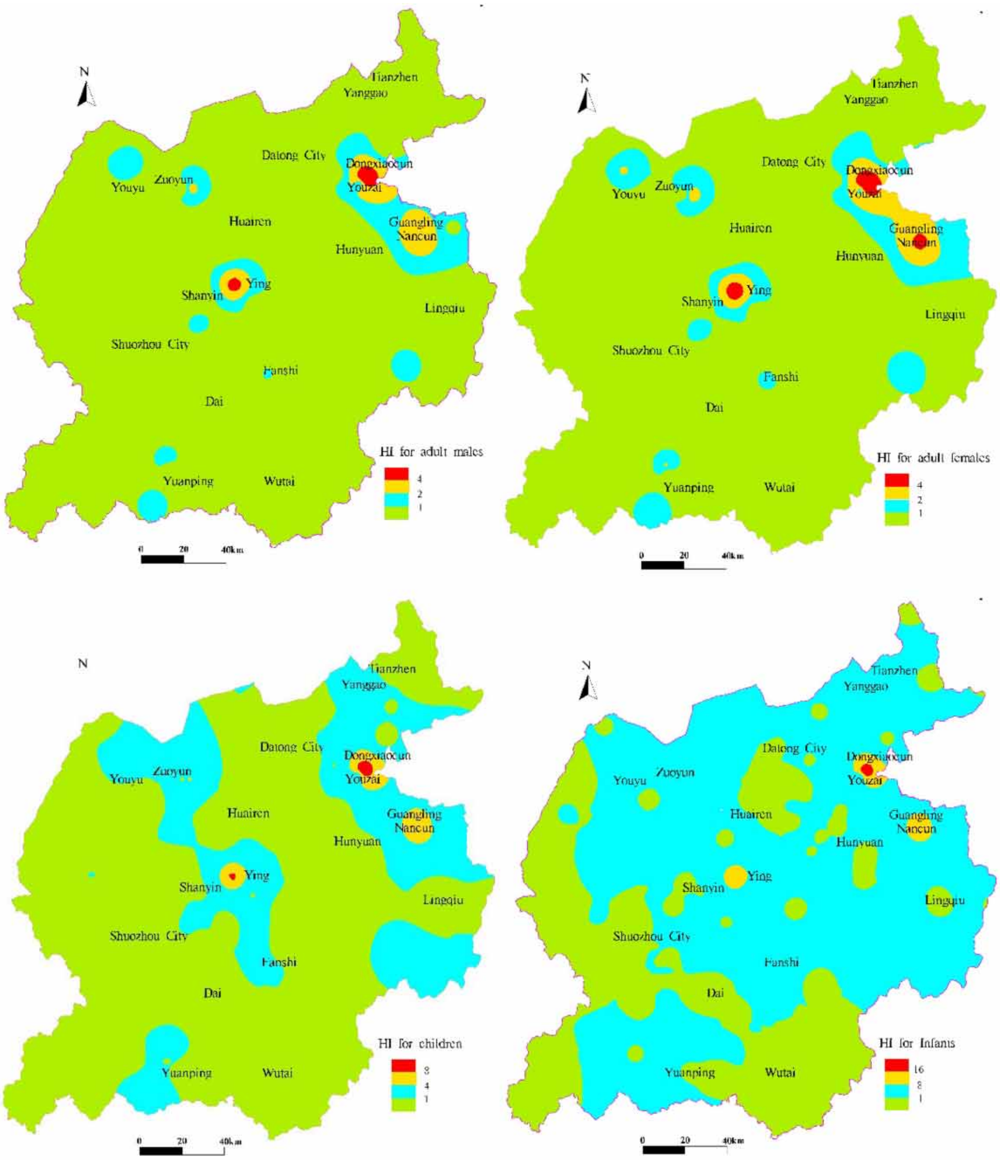


Figure 7 | Health risk distribution map.

Dongxiao village and Nancun village. There is a health risk of NO_3^- pollution, which is basically the same as that of adult males. The results showed that NO_3^- pollution of groundwater in the study area did little harm to the adult population; however, about 10% of the study area belongs to the health risk area, which is located in the middle and northeast of the study area. Adult female risk areas (HI $<$ 1) roughly overlap with male risk areas (Figure 7). However, the maximum HI for women and men were 9.84 and 8.55, respectively. Therefore, compared with adult women, adult men have a lower risk, which is due to physiological factors. This finding is consistent with that of Weining plain in Northwest China, and emphasizes the importance of gender in the process of HHRA.

HEALTH RISKS FOR INFANTS AND CHILDREN

The health risk calculation results of children and infants are shown in Figure 7. Low-risk areas for infants are distributed around Shuozhou City and Datong City, while high-risk areas are distributed in Yingxian County, Dongxiao village, South village, Zuozhi and Fanshi. The high-risk area accounts for 39%, indicating that 39% of infants' HI is unacceptably high. The HI value in this area is between 1.01 and 25.32, indicating that infants in this area have a high potential health risk. For children, 83% of the study areas had acceptable HI levels distributed in Shuozhou City, Datong City and most of the surrounding areas. The HI of 17% of children in the study area is unacceptably high. The HI value in this area is between 1.08 and 14.21, distributed in Yingxian County, Dongxiao village, Nancun and other areas, indicating that children in this area have high potential health risks. The HI value of children is lower than that of infants, and the maximum value is 14.21. The total area of health risk area is 21% less than that of infants.

The above results show that the risk of NO_3^- in infants in the Datong Basin is higher than that of minors and adults, and the risk of women is higher than that of men, and infants need to be protected more, which is related to the difference in weight of different age groups. The results are consistent with those of Chen *et al.* (2005). The risk ratio of different populations is shown in Figure 8. The risk in urban areas is lower than that in rural areas, mainly due to the use of NO_3^- fertilizer and organic fertilizer in rural areas, and cities are not affected by NO_3^- fertilizer and organic fertilizer.

DISCUSSION

It can be seen from Figures 1, 4 and 6 that in the alluvial-proluvial plain area with low altitude and small topographic gradient, agricultural production activities are relatively developed, and the amount of fertilizer and other fertilizers used in farmland is large. Farmland irrigation and rainfall in plain areas are the main recharge sources of groundwater (Amir *et al.* 2021; Zhang

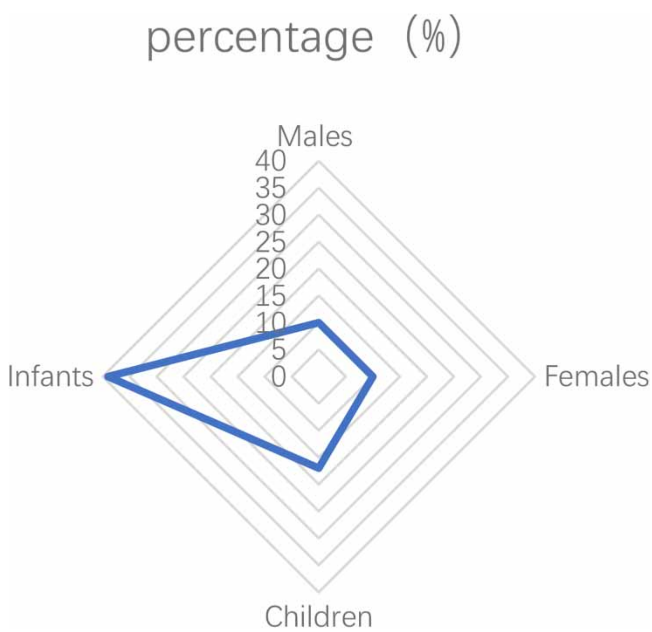


Figure 8 | Risk proportion chart of different categories of people.

et al. 2021). After farmland irrigation or rainfall, most of the water, except for partial evaporation and absorption by plants, seeps into the aeration zone or groundwater. The nitrogen components contained in fertilizer will gradually enter the groundwater environment with the infiltration of groundwater, resulting in nitrate pollution in groundwater (Anju & Kavita 2022). Therefore, compared with the mountainous areas with higher altitude and larger topographic gradient, the groundwater in the alluvial-proluvial plain areas with lower altitude and smaller topographic gradient is more vulnerable to agricultural non-point source pollution (Marghade *et al.* 2021; Nitika *et al.* 2021).

Agricultural non-point source pollution leads to the continuous accumulation of nitrate in groundwater, and high nitrate concentration will cause serious harm to human health. Hypermethemoglobinemia is a health problem caused by high nitrate in drinking water. When the nitrate content is high in the body of newborns, cyanosis will also occur, which will lead to mucous membrane blueness and digestive and respiratory diseases in infants (Saravanan *et al.* 2022). The intake of large amounts of nitrate has a complex relationship with cancer, adverse reproductive outcomes and other health problems, which requires further research in the future. In this paper, the health risk assessment of adult men, adult women, children and infants is carried out respectively, and the corresponding conclusions are drawn, which is conducive to improving people's awareness of the protection of infants and children against nitrate risk.

Abooalfazl *et al.* (2019) evaluated fluoride and nitrate pollutants in drinking water resources and their health risks in semi-arid areas of southwestern Iran. This study showed that children, adolescents and adults are vulnerable groups as they are exposed to high concentrations of fluoride and nitrate in drinking water, posing a non-carcinogenic risk. Alimohammadi *et al.* (2018) conducted a determination and risk assessment of nitrate concentration in bottled water in Iran. The results showed that the nitrate concentration in bottled water ranged from 0.146 to 0.1 mg/L (average 10.55 mg/L), with one type of bottled water having a higher nitrate content than the World Health Organization guidelines. Radfard *et al.* (2019) conducted a health risk assessment of fluoride and nitrate in drinking water for rural residents in the arid southeastern region of Bardaskan, Iran, and found that the highest and lowest concentrations of nitrate and fluoride occurred in the northeast region. These studies indicate that the health risk of nitrate pollution is a global health issue that needs to be given sufficient attention.

Han *et al.* (2021) conducted a health risk assessment model on four groups of people: infants, children, adult males and adult females. The results showed that long-term consumption or exposure to groundwater with high nitrate levels in the area would pose great risks to infants and children. Tian *et al.* (2020a, 2020b, 2020c, 2020d) conducted a study on the health risks of groundwater chemistry and nitrate intake in Helen City, and found that human activities are the cause of the formation of chemical composition in shallow groundwater in Helen. The degree of harm caused by NO_3^- pollution is in the order of infants > children > adult females > adult males. The above two conclusions are consistent with this study, and the research results can provide technical reference and theoretical support for the evaluation of groundwater nitrate under the same type of hydrogeological conditions.

The research results of predecessors, as well as the experience and test data obtained from this survey, provide strong support for this study. However, there is limited experimental research on the impact of nitrate pollution on human health, which points out the research direction for future scientific research.

Dynamic monitoring of groundwater indicators, timely detection of changes in pollutants, scientific assessment of pollutant risks, precise implementation of policies and ensuring drinking water safety are suggested.

CONCLUSION

Through research, qualitative and quantitative conclusions have been drawn on the current situation of water quality, influencing factors and NO_3^- exceeding the standard in the research area. Qualitative and quantitative conclusions have also been drawn on the health risks caused by NO_3^- exceeding the standard. These new achievements will help people improve their awareness of risk prevention, protect the environment, respect nature, implement precise policies and develop scientifically.

Research has found that the groundwater in the study area is mainly classified as Class II water, with overall good quality. Although Class V water has not been identified, Class IV water accounts for a relatively large proportion and requires strengthened monitoring and remediation. The main influencing factors for water quality are NO_3^-, Fe, F^- and SO_4^{2-}.

About 34% of NO_3^- samples in the study area exceed the Class III value, and the use of agricultural fertilizer is the main source of NO_3^- in the groundwater in the area.

There are significant differences in the health hazards of groundwater NO_3^- pollution on different populations, and infants and children are more vulnerable to nitrate pollution.

The division of pollution areas and high-risk groups has an important guiding role for health risk prevention. In the polluted area, people can distinguish people of different sexes and ages for scientific protection.

NO_3^- is significantly correlated with $CaCO_3$, Cl^- and SO_4^{2-}, and the increase in NO_3^- concentration is related to the use of NO_3^- fertilizers, organic fertilizers and the discharge of domestic sewage in agricultural production.

In the future, we should further research and develop simulation and evaluation methods to improve the accuracy and reliability of prediction and evaluation, in order to protect the sustainable use and management of groundwater resources.

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AVAILABILITY OF DATA AND MATERIAL

The data and material data provided by the laboratory of *Harbin Center of Natural Resources Comprehensive Survey, CGS* are reliable.

DATA AVAILABILITY STATEMENT

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

CONFLICT OF INTEREST

The authors declare there is no conflict.

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