

# Investigation of quality and pollution characteristics of groundwater in the Hutuo River Alluvial Plain, North China Plain

Yasong Li<sup>1,2</sup> · Zhaoji Zhang<sup>1</sup> · Yuhong Fei<sup>1</sup> · Honghan Chen<sup>2</sup> · Yong Qian<sup>1,2</sup> · Yu Dun<sup>1</sup>

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**Abstract** Groundwater plays a critical role in northern China, accounting for 62 % of the total water supply in the Beijing–Tianjin–Tangshan region. Groundwater is particularly important in the North China Plain—the largest alluvial plain and one of the most densely populated regions of East Asia. The Hutuo River Plain, which is located in the western portion of the North China Plain, is essential in supplying groundwater to the entire central region of the plain. Groundwater in this region is the main source of industrial, agricultural, and drinking water and therefore directly influences daily life. In this study, 482 groundwater samples including 390 shallow groundwater samples and 92 deep groundwater samples were collected and analyzed to investigate water quality and the effects of anthropogenic activities in the Hutuo River Plain. Application of the improved fuzzy mathematics evaluation method revealed that 21.5 % of the samples exceeded the Class III water standard, and that these samples were primarily in areas containing high concentrations of industrial activity or sewage discharge, and at the same time the native hydrogeological environment is the major factor influencing distribution of most pollution components, groundwater quality in the top of the Hutuo River alluvial–proluvial fan was not very good just as the Vadose zone lithology is a little more coarse, in the leading edge of the alluvial–proluvial fan the vadose zone is mainly formed of

clay with very low vertical permeability, these characteristics endow the area with good protective performance, which makes it difficult for surface pollutants to enter the aquifer. Groundwater quality in the region was primarily affected by inorganic materials including total hardness, iron, manganese, total dissolved solids, and nitrate nitrogen, the rates of exceed Class III water standard was 26.45, 17.72, 11.11, 10.58, and 8.47 %, respectively, in shallow groundwater, and 20.20, 16.34, 16.35, 14.43, 2.52 %, respectively, in deep groundwater. There are several organic materials with high detection rates including chloroform, carbon tetrachloride, carbon dichloride, and ortho-dichlorobenzene, the detection rate was 14.02, 2.12, 3.44, 2.48 %, respectively, in shallow groundwater, and there were little organic compounds detected in deep groundwater. Overall, the results indicated that the hydrogeological environment and anthropogenic activities have led to the introduction of toxic metals, nitrogen, and organic materials into groundwater in the Hutuo River Plain. These findings will be useful to future studies of the region and will facilitate the remediation and prevention of contamination in the area.

**Keywords** Hydrogeology · Groundwater · Fuzzy mathematics · Quality evaluation · Hutuo River Plain · Pollution factor

✉ Yu Dun  
dunyu2005@gmail.com

<sup>1</sup> Institute of Hydrogeology and Environmental Geology, Chinese Academy of Geological Sciences, No. 268, Zhonghuabei Street, Shijiazhuang 050061, Hebei, China

<sup>2</sup> School of Water Resources and Environment, China University of Geosciences, Beijing 100083, China

## Introduction

Groundwater is not only a primary source of freshwater, but also a sensitive component of complex eco-environmental systems. Accordingly, fluctuations in groundwater characteristics tend to affect the natural balance of the local ecosystem. Water quality is very important to human

health, as well as the quantity and quality of crops via its effects on soils and the environment (Hoek et al. 2001; Kirda 1997). The North China Plain (NCP) is the largest alluvial plain and one of the most densely populated regions of East Asia. The population, economic activity, and agricultural production of the area have greatly increased over the last few decades, resulting in increased water demand (Foster et al. 2004). Groundwater plays a critical role in northern China, accounting for 62 % of the total water supply in the Beijing-Tianjin-Tangshan region (BTT) (Nie et al. 2011). Once becoming polluted, contaminated groundwater would show the potential to harm human health directly or indirectly (Wang and Shang 2002). Human activities in the NCP have increased while the amount of precipitation has decreased, which may have led to a decrease in shallow groundwater recharge (Wang et al. 2010). Under such conditions, the quality of water would be especially important.

During the early stage, conventional components and toxic heavy metals were the major groundwater pollution sources. However, over the last 30 years, abstraction by industry and the widespread use of organic reagents and organic products have resulted in toxic and organic pollution of water becoming a new research focus (Jiang 2003; Guzzella et al. 2006). Such organic pollutants in groundwater can impact human health to varying degrees (Shi et al. 2000; Li et al. 2010; Gao and Wand 2011). To date, 2221 types of organic chemical pollutants have been detected in aquatic systems worldwide, among which mononuclear aromatics, halohydrocarbons, and organic-chlorine pesticide have appeared in groundwater in China (Guo et al. 2006).

Historically, groundwater in the Hutuo River Plain originates from the Hutuo River, which is one of the major rivers in the NCP that originates from Tai Hang Mountain. The Huto River joins the Fu Yang River and eventually drains into the Bohai Sea.

The Hutuo River Plain, which is located in the western portion of the North China Plain (Fig. 1), is in the piedmont recharge area; therefore, groundwater quality in this region is of vital importance to the entire central region of the NCP. Accordingly, pilot studies and evaluations of the pollution conditions of the Hutuo River Alluvial Plain are important to investigations of groundwater quality and overall pollution conditions in this area.

Agricultural production, population and industrial and mining enterprises have been increasing in the study area since the 1980s, which has resulted in increased demand for water resources and therefore increased groundwater abstraction. This has resulted in continuous lowering of the groundwater table, as well as the formation of regional groundwater depression cones, which have given rise to a

series of environmental and geological problems including deterioration of water quality and land subsidence (Bi et al. 2001). Since factories are usually located near rivers, the majority of industrial wastewater and domestic sewage is discharged into rivers through sewer systems. However, these systems often do not have leakage prevention, resulting in direct or indirect infiltration of wastewater into local aquifers and subsequent contamination of the groundwater resources.

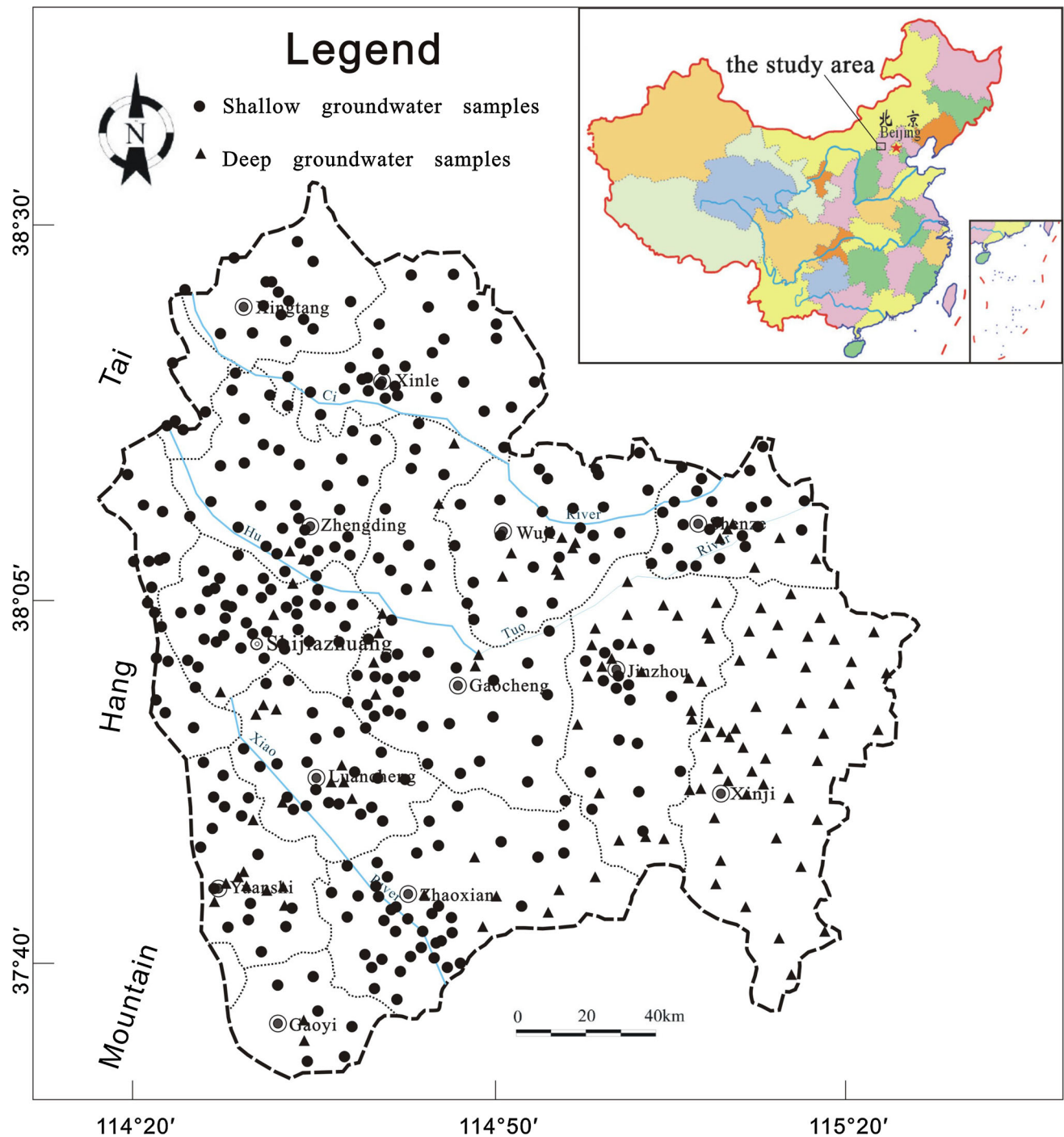
The aim of this study is to know the groundwater quality condition in this whole region, and to find out the typical compounds in groundwater especially the organic compounds in groundwater, as this is the first time to test the organic compounds for the whole study region, and to find out what's the main pollution sources to influence the groundwater quality.

## Data sources and research methods

### Hydrological setting of the research area

The eastern part of the research area is connected to the alluvial plain in the river course with the north-northeast trend, the northern and the southern parts are both flanks of the alluvial fan, the development of the water-bearing level is gradually worsen, the water-bearing level represents the inter-fan low land phase accumulation, and the depth of the quaternary loose deposits is increased to 600 m in the east part from 200 m in the piedmont. The west part of the aquifer is a thick layer of sandy gravel, and this material becomes finer to the east, eventually transitioning to primarily coarse sand. Medium fine sand and powder fine sand are commonly distributed in the transitional zone between the eastern part of the site and the fan edge, and fine sand mainly distributed in the partial zone. The overall underground flow is from northwest to southeast, the main compensation mode in groundwater is atmospheric precipitation, infiltration of river and channel water, and irrigation return water, and drainage primarily occurs via manual exploitation.

The aquifer in this area is primarily composed of Quaternary cobbles, gravel, sand, and laminated or lensed clay to a depth of 400–500 m. The aquifer is separated into four groups: holocene ( $Q_4$ , up to 10 m thick), upper Pleistocene ( $Q_3$ , 20–70 m thick), middle Pleistocene ( $Q_2$ , up to 20 m thick) and lower Pleistocene ( $Q_1$ , up to 30 m thick).  $Q_3$  is the major stratum for supply of domestic and agricultural water in the area that is subject to pollution due to its relatively high hydraulic conductivity ( $k = 27.5\text{--}70.2$  m/d) (Chen et al. 2005; Yang et al. 2001).



**Fig. 1** Location of study area and groundwater samples

**Sampling procedure**

From August 11 to November 30 of 2009, 390 groups of shallow groundwater and 92 deep groundwater samples were sampled in the research area with a sampling density 3–4 groups per km<sup>2</sup> (Fig. 1). Strict quality control was conducted during the sampling, sample delivery and testing

process. The sampling equipment is an independently developed groundwater sampler, which is made up of steel pipes and polytetrafluoroethylene hoses. Wells were cleaned before sampling, and the electric conductivity (EC), water temperature (*T*), and the pH of the water was measured in situ using a portable analyzer (WTW-Muti340i, Germany). During sampling, the sample bypass valve was

adjusted to stabilize the sampling flow rate at 0.2–0.5 L/min. Collected samples were stored in a refrigerator (4 °C) until delivery to the Groundwater Mineral Water and Environmental Supervising and Testing Center of the Ministry of Land and Resources for testing and analysis. Samples were analyzed in parallel by ALS Chemex (Shanghai) Co. Ltd. All samples were analyzed within 4 days by combined purge and trap gas chromatography mass spectrometry.

### Methods of groundwater quality assessment

Multiple water quality evaluation methods and models, including the comprehensive index method, fuzzy comprehensive evaluation method, and the gray clustering method, have been developed and approved to be powerful in water quality assessment (Li et al. 2014; Yusuf 2007; Alexander 2008), especially the fuzzy comprehensive evaluation method have been studied by lots researchers (Li 2009; Miao et al. 2014; Xu et al. 2014; Zhang et al. 2014). Although each of which have advantages and disadvantages (Li et al. 2011; Li et al. 2012a). The improved fuzzy mathematical comprehensive evaluation method was employed in this study, making analysis of the quality of groundwater comparatively objective.

In the conventional sense, the fuzzy mathematical evaluation method employs a fuzzy line to describe the groundwater quality by means of subordinate degree based on the fuzzy transformation principle. The matrix formed by the subordinate degree of each individual index to the corresponding class of water quality is called the fuzzy relation matrix. The weight matrices of the evaluating indices are then formed based on the physicochemical properties and the toxicological characteristics. Combining the four operators (the min–max algorithm, multiplying–max algorithm, min–adding algorithm and multiplying–adding) yields the comprehensive evaluation results of the groundwater quality. When there are multiple evaluation

indices, especially trace indices such as the heavy metal index and the organic index, the taking of the operator of multiplying–adding will be more objective (Li et al. 2009). However, the evaluation results might be easily influenced by the over-limit indices in the conventional groups. For example, some water samples may be overrated as Class II or Class III because there is only one parameter such as heavy metal or organic component that exceeds the limit, regardless the toxicology of the heavy metal and organic components which could not be ignored. Moreover, this part of the evaluation may result in improper utilization of the groundwater resources. Improvements in the evaluation method have been made to evaluate the groundwater quality objectively and guide people to effectively use groundwater resources. Such efforts have focused on modification of the evaluation results for samples having toxicological indices exceeding the limit that are still rated as Class I–III being changed to be Class IV. Accordingly, the revised evaluation results can reflect the quality of the groundwater objectively.

## Results and analysis

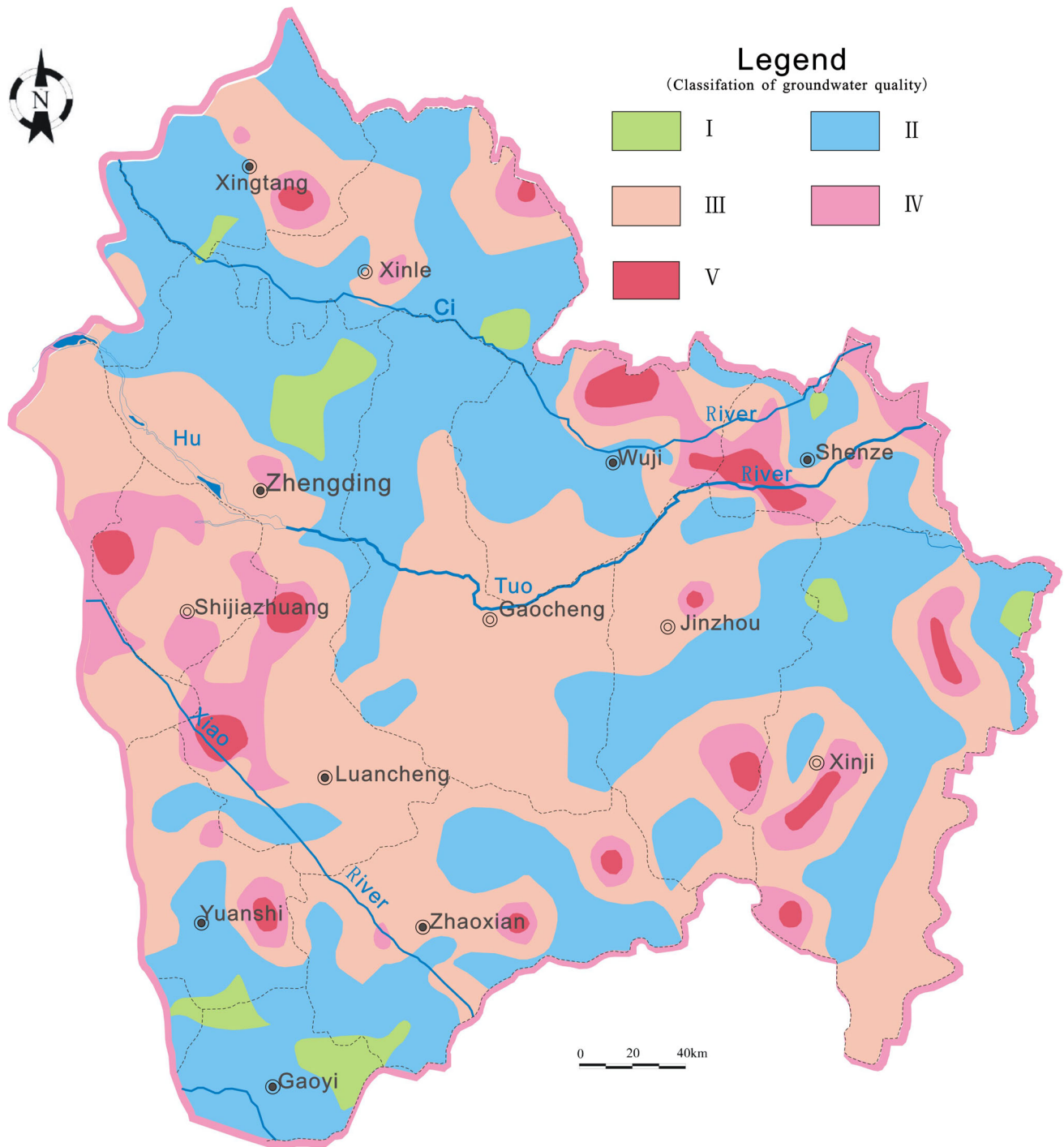
### Groundwater quality

A total of 52 hydrophysical and hydrochemical indices (shown in Table 1) were selected for comprehensive assessment of groundwater qualities.

The comprehensive evaluation results (Fig. 2) shows that the sampling points with water quality exceeding Class III comprised 21.5 % of the total sampling points, and that most were mainly distributed in three areas. (1) Southeast downtown Shijiazhuang, which is an old industrial base and the center of several other industries. This region contains the North China Pharmaceutical Co., Ltd., the Shijiazhuang Coking Plant, the Third Cotton Spinnery, the Sixth Cotton Spinnery and several other major enterprises

**Table 1** System of evaluation indices

Classification	Organic indicators	Inorganic indicators
Conventional indicators	Trichloromethane, carbon tetrachloride	pH, Fe, Mn, Cu, Zn, Al, Na, chloride, sulfate, total hardness, TDS, COD, As, Cd, Cr, Pb, Hg, Se, fluoride, nitrate, ammonia nitrogen, iodide
Unconventional indicators	1,1,1-trichloroethane, trichloroethylene, tetrachloroethylene, dichloromethane, sym-dichloroethane, 1,1,2-trichloroethane, 1,2-dichloropropane, tribromomethane, vinyl chloride, 1,1-dichloroethane, sym-dichloroethane, chlorobenzene, <i>o</i> -dichlorobenzene, <i>p</i> -dichlorobenzene, benzene, methylbenzene, para-xylene, ethylbenzene, styrene, hexachlorocyclohexane, $\gamma$ -hexachlorocyclohexane, dichlorodiphenyltrichloroethane, hexachlorobenzene, benzo(a)pyrene	Ba, Mo, Ni

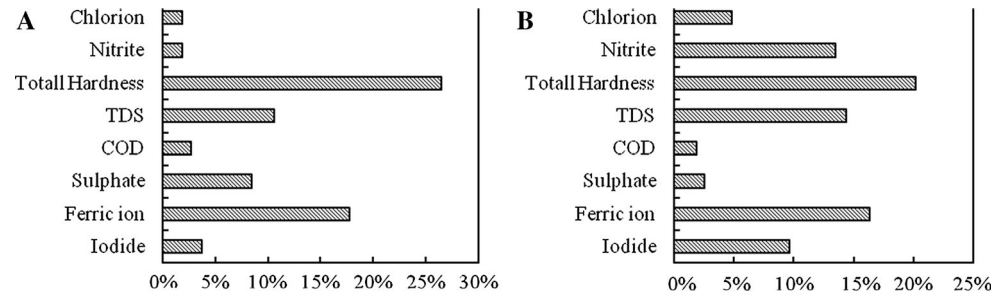


**Fig. 2** Groundwater quality evaluation results

concentrated on the south side of the Shijiazhuang-Taiyuan railway. These facilities are the major factors influencing the groundwater quality of this area. (2) The area along the sewage discharge channel. The Cihe River, Mudao Ditch, Hutuo River and Jiaohe River in this region have all become wastewater drains for nearby factories. Lateral leakage has a large impact on shallow groundwater, while

sewage irrigation in some areas has enlarged the impact area. (3) The area where aquifer show high groundwater vulnerability. The Hutuo River Alluvial Plain is located in the axis of the top of the alluvial-proluvial fan and contains a vadose zone composed of sandy soil. Although the structure and lithology of the vadose zone are advantageous for groundwater recharge, they result in

**Fig. 3** Comparison of exceeding-standard rates of inorganic components. **a** Shallow groundwater samples; **b** deep groundwater samples



comparatively poor pollution resistance. As a result, the industrial sewage discharge, infiltration of landfill leachate and seepage of sewers lead to groundwater contamination.

### Exceed standard of inorganic components

Exceed standard ratio of the total hardness, Fe, Mn, iodide, total dissolved solids, sulfate ion and nitrate were relatively high (Fig. 3). The over-limit ratio of Fe and Mn may be caused by the natural environment, while that of the total hardness and total dissolved solids was likely caused by anthropogenic activities. In addition to the landfill leachate and sewage infiltration from the polluted river, human activities in this region also include the excessive pumping of shallow groundwater, which leads to the formation of groundwater depression cones and thickening of the vadose zone. The excessive pumping of groundwater may also cause changes in the hydrodynamic field and the hydrogeochemical environment, as well as hydrogeochemical interactions between pollutant carriers, the vadose zone and the water-containing surrounding rocks as well. Under these conditions, the soluble and insoluble calcium and magnesium salt in the soil and sediment of the under layer and the exchangeable calcium and magnesium will transfer from the solid phase into the water, increasing groundwater hardness and the total dissolved solids.

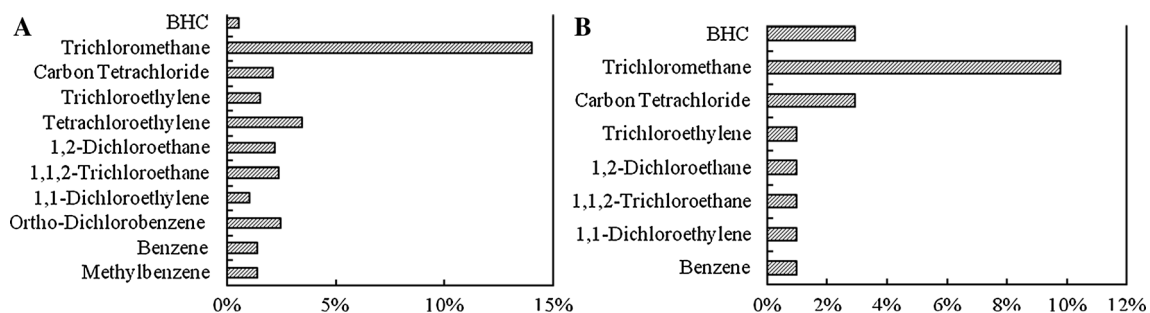
In research areas with poor anti-pollution capacity, pollution caused by nitrogen pollutants mainly occurs in shallow groundwater. This is reflected by a relatively high over standard ratio of nitrate nitrogen, with most of the over standard ratio points being located in the axis of the

top of the Hutuo River alluvial-proluvial fan, where the vadose zone is mainly formed of sandy soil with poor pollution resistance. In this area, industrial waste discharge, nitrogenous fertilizer, and household landfill materials may lead to excessively high levels of the three types of nitrogen components. In several areas, nitrite nitrogen in groundwater exceeded the standard, which likely means that there has new contamination accident happened on the surface.

Evaluation of toxic heavy metals in the study area showed that lead and chromium VI had a high detection rate (67.05 and 14.35 %, respectively), even though they were only detected in a few areas. Conversely, almost no arsenic, cadmium or mercury were detected. The detection of toxic heavy metal is greatly affected by the distribution of landfills and factory discharge conditions.

### Detection of organic components

A total of 22 types of organic components have been detected in this area, among which halohydrocarbon (including trichloromethane, tetrachloroethylene and carbon tetrachloride) and *o*-dichlorobenzene (Fig. 4) have relatively high detection rates. The detection rate of trichloromethane is 13.13 %, while that of tetrachloroethylene and carbon tetrachloride are comparatively low (2.71 and 2.29 %, respectively; Table 2). Additionally, a few other components such as trichloroethylene, 1,2-dichloroethane and *o*-dichlorobenzene were also detected, with detection rates of about 1.5 %. Semi volatile components showed a generally low detection rate, with the total BCH detected



**Fig. 4** Comparison of detection rates for organic components. **a** Shallow groundwater samples; **b** deep groundwater samples

**Table 2** Organic pollution characteristics

Compound	Detection limit (µg/L)	Detection rate (%)	Average detection concentration (µg/L)	Class III water standard (µg/L)	Over-limit ratio (%)	Average over-limit concentration (µg/L)
Trichloromethane	0.5	14.02	2.96	60	0	0
Tetrachloroethylene	0.5	3.44	2.29	40	0	0
<i>o</i> -Dichlorobenzene	0.5	2.38	2.92	1000	0	0
1,1,2-Trichloroethane	0.5	2.38	13.20	5	0.26	109.6
1,2-Dichloroethane	0.5	2.12	1.00	30	0	0
Carbon tetrachloride	0.5	2.12	7.91	2	0.53	28.2

**Table 3** Pollution indices of halogenated hydrocarbons in soils and sediments

Compound	Chloroform	Carbon tetrachloride	1,1,2-Trichloroethane	Tetrachloroethylene	1,2-Dichloroethane	1,2-Dichlorobenzene
Soils	4.71	4.90	4.19	4.00	4.24	2.56
Sediment	6.31	6.41	5.61	5.29	5.78	3.45

from five groups of water samples and total DDT from one group of water samples. The variations in detection and amounts in shallow groundwater were higher than those in deep wells. The main components detected in the shallow groundwater include trichloromethane, tetrachloroethylene, carbon tetrachloride, and 1,1,2-trichloroethane. Comparatively, no tetrachloroethylene was detected in deep groundwater, while other components showed lower detection rates. Throughout the region, the halohydrocarbon and chlorobenzene pollutants were the main sources of organic pollution, while benzene, mononuclear aromatics and polycyclic aromatic hydrocarbons were present at relatively low levels or absent (Li et al. 2012b) (Table 3).

Organic pollution components in the research area mainly originated from human activities rather than the natural environment. Most regions polluted with organic compounds are located in industrial areas or near sewage discharged rivers.

**Discussion**

The Hutuo River alluvial area is near a piedmont recharge area in which the vadose zone is formed of relatively coarse particles, resulting in relative high groundwater recharge rate and relative quick groundwater refresh cycle. As a result, this area has long been characterized by shallow groundwater with good quality. However, increased urbanization, industry and agricultural activity have resulted in the groundwater becoming contaminated to different degrees, and groundwater quality in the research area is deteriorating obviously. This study was conducted to identify factors that

influence the pollution conditions and contribute to groundwater pollutants.

**The native hydrogeological environment is the major factor influencing distribution of most pollution components**

The local native hydrogeological environment has a restrictive function on the occurrence and migration of pollution components. The top of the Hutuo River alluvial-proluvial fan contains the cities of Shijiazhuang and Xinle, as well as Luancheng County. The vadose zone in this area shows coarse lithology and good permeability, which make it easy for pollutants on the surface to enter the aquifer. Nitrogen pollutants are concentrated here, and there is a high detection rate of organic components in many indices. The City of Gaocheng and Wuji County are located in the center of the alluvial-proluvial fan, where the surface layer contains particles that change from coarseness to fineness, with increasing clayey soil interlayers and decreasing permeability. As a result, pollution decreases in a vertical direction, inorganic over-limit areas have only been detected in cities and towns or along the sides of sewage rivers, and organic components have been detected in only a few spots. In the leading edge of the alluvial-proluvial fan, which contains the cities of Shenzhe and Xinji, the vadose zone is mainly formed of clay with very low vertical permeability. These characteristics endow the area with good protective performance, which makes it difficult for surface pollutants to enter the aquifer. Additionally, the concentration of the lateral pollutants becomes very low when reaching the edge of the fan via the transitional zone, which can hold back and absorb some of the pollutants.

### **Increased anthropogenic activity is the main cause of the detection and over-limit of toxic heavy metals, the three types of nitrogen pollutants and organic pollutants**

Since the late 1970s, the Hutuo River Plain has been subject to a rapidly growing economy, increased urbanization and expanding population, so that groundwater pollution has increased. Conducting a general survey of areas with inferior groundwater revealed that these areas are primarily characterized by large populations, intensive industrial activity and frequent human activity. Generally speaking, the intensity of human activity is directly related to the quality of groundwater.

### **Seriously polluted surface water poses an immediate threat to groundwater quality**

In the research area, most of the rivers are seasonal streams influenced by upstream water conservancy projects and the arid climate. As a result, the majority of rivers are dry year-round, although some near the piedmont receive runoff for a short time during flood season. In addition, most rivers on the edge of the alluvial-proluvial fan become sewage discharge channels for cities and towns and industrial and mining enterprises on their banks. Industrial wastewater is discharged into nearby sewage channels directly, while domestic sewage enters sewage channels via the sewer. As a result, the majority of rivers in the area are seriously polluted. Indeed, among the 62 monitoring sections of rivers in the North China Plain, 48 sections meet the Class V water standard, while only eight meet the Class III water standard (China surface water environmental quality standard, GB\_3838-2002). In the research area, many rivers are severely polluted, including the Hutuo, Jiaohe, and Mudao rivers. In addition to the conventional ionic and toxic heavy metals detected in the rivers, the organic components are also detected and some even exceed the limit, which include the benzene, ethylbenzene, methylbenzene, dimethylbenzene, dichloromethane, 1,2-dichloroethane, trichloromethane, Benzo [a] pyrene, etc. The pollutants in river water infiltrating into the soil tend to pose an immediate threat to the quality of shallow groundwater.

### **Long-term sewage irrigation has strong effects on soil and groundwater**

The sewage irrigation area (about 438.8 km<sup>2</sup>) is located along Dongming Ditch and the Jiaohe River, which receives annual sewage of 304 million m<sup>3</sup>. The major sources of irrigation water in the region are industrial wastewater and domestic sewage, which contain contaminants such as chloride, COD<sub>Mn</sub>, BOD<sub>5</sub>, SS, and sulfide.

Farms in Luancheng, Yuanshi, and Zhaoxian counties are irrigated with sewage throughout the year, resulting in decreasing soil fertility, reduced crop output and incalculable damage to the shallow groundwater.

### **Low processing rate of industrial and domestic sewage together with the mass discharge of wastewater in the rural–urban continuum cause pollution to the soil and groundwater**

The Hutuo River alluvial-proluvial fan contains more than 800 enterprises, which has resulted in a low processing rate of industrial and domestic sewage and disorganized discharge of waste streams. Moreover, a field survey revealed that several township enterprises were discharging unprocessed industrial sewage without permission, exacerbating pollution in the region.

### **Industrial and domestic refuse landfills cause severe soil and groundwater pollution**

Widely scattered domestic waste in urban areas include household refuse, waste plastics, paper, metal and coal ash, which contain sulfate, chloride, ammonia impurities and organic materials. During biodegradation and rainwater leaching, these materials may generate Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>, COD, Mn and filtrate with a high level of suspended solids. Accordingly, the random disposal of such wastes without any anti-seepage measures can lead to groundwater contamination.

### **Overuse of chemical fertilizer and pesticides in agriculture have direct impacts on the quality of soil and groundwater**

A total of 1.70 million tons (physical quantity) of chemical fertilizer and 130 million tons of pesticides are used in the Hutuo River Plain annually. The excess chemical fertilizers and pesticides seep into the soil and groundwater via surface water runoff from farms, drainage and underground seepage during rainfall or irrigation processes, resulting in contamination. After entering a water body, chemical fertilizers and pesticides diffuse, resulting in expanded pollution range and extensive water pollution; accordingly, these materials pose a serious threat to groundwater quality.

## **Conclusions**

Overall, the results of the present study indicate the following:



- (1) Evaluation of the water quality revealed that areas with water exceeding Class III of the pollution standard comprise 21.5 % of the total sampling points, which are mainly located in industrial distribution centers and areas along sewage discharged river. Additionally, the local hydrogeological conditions impact the quality of groundwater to some extent, which is reflected by the fact that water quality in the shaft of the alluvial-proluvial fan is worse than that in the center and edge of the fan.
- (2) In the research area, inorganic components with a high detection rate include total hardness, Fe, Mn, iodide, total dissolved solids, sulfate ion and the nitrate, while organic components include halohydrocarbons such as trichloromethane, tetrachloroethylene and carbon tetrachloride, as well as *o*-dichlorobenzene. Components with a high detection rate also have a high pollution index value, indicating good consistency.
- (3) The chemical index detection conditions and analysis of the over-limit factors of the groundwater indicate that the native hydrogeological environment and anthropogenic activities are the major factors responsible for the presence of toxic metals, nitrogen pollutants and organic pollutants. Surface water contamination, sewage irrigation, waste stacking and overuse of chemical fertilizers and pesticides are the root causes of groundwater pollution.

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