Spatio-temporal variation of groundwater contamination using IEA-UEF in urban areas of Jilin City, North-eastern China

Zhang Nan, Liu Bo and Xiao Changlai

ABSTRACT

Groundwater monitoring wells located in urban areas of Jilin City were sampled from 1980 to 2009 for eight groundwater quality parameters: pH, SO$_4^{2-}$, Cl$^-$, NO$_3^-$-N, NO$_2^-$-N, NH$_4^+$-N, F$^-$, and total hardness (TH). The data were analysed by a universal exponential formula based on an immune evolutionary algorithm, and later mapped with the Kriging interpolation method. The primary objectives were to assess the main parameters that influenced groundwater quality and the spatio-temporal variability of groundwater contamination over several years. The results showed that NO$_3^-$, NH$_4^+$, and TH were the main parameters that influenced groundwater quality. Spatially, groundwater was polluted in all urban areas to varying degrees, and the Jiangbei district was the most heavily polluted location. Temporally, the groundwater contamination status could generally be classified into four stages and showed the following pattern during 1980–2009: heavy – light – heavy – light.

Key words | groundwater contamination index, immune evolutionary algorithm, Jilin City, spatio-temporal evolution, universal exponential formula

INTRODUCTION

Groundwater availability and quality greatly influence the environment, economic growth, and the health of nearby residents. Industrialization and urbanization, together with intensified agricultural activity associated with an ever-increasing population, have given rise to increased water demands on one hand, and the potential for large-scale emissions of contaminants to shallow groundwater, a source of drinking water, on the other hand (Joarder et al. 2008). Meanwhile, the deterioration of groundwater quality due to human activities can also cause adverse effects on human health and the natural ecosystem. Almost half of the population in developing countries are subject to health threats deriving from a lack of drinking water or the presence of microbiologically contaminated water (Van Leeuwen 2000). Therefore, a safe water supply, including groundwater and surface water, must be guaranteed.

Groundwater present in the urban areas of Jilin City comprise about 20% of the total water supply. The drinking water supply is mainly served by the Second Songhua River, and suffers from an over-dependency on this source. If a large and heavy pollution accident occurred in the Songhua River, such as the contamination incident involving the Jilin Petro-Chemical Co. Ltd in 2005, it would threaten the normal production and lives of residents, as well as societal stability. After the 2005 contamination incident, the related decision-making department of Jilin City has set out to establish emergency groundwater resources. The clarification of spatio-temporal characteristics of groundwater quality in urban areas of Jilin City is the key task required for completion of the above project. This spatio-temporal heterogeneity of shallow groundwater results from the cumulative effects of a large number of variables that may be impossible to isolate and study individually. These are hydrometeorology, topography, drainage system efficiency, anthropogenic activities, land use, and the associated chemical concentrations in...
topsoil, net vertical recharge (affected by leaching rainfall), local depth to groundwater, and lateral recharge from ground or surface water sources, and their associated impacts (Kim et al. 2005; Muñoz-Carpena et al. 2005; Rouxel et al. 2011; Simpson et al. 2011).

The aim of this study is to investigate and characterize dynamic changes in the quality of shallow groundwater in urban areas of Jilin City in past years, using a universal exponential formula (Li et al. 2004, 2008). Earlier studies in this region have noted some of these changes (He & Li 1987; Liang et al. 2007; Wu et al. 2012; Wei et al. 2014); however, they have lacked comprehensive analyses or investigations that utilized the aforementioned analytical method. The methods used in past groundwater quality evaluations generally combined multivariate statistical methods and geostatistical modelling (e.g. Helsrup et al. 2007; Papatheodorou et al. 2007; Cloutier et al. 2008; Li & Zhang 2008; Dassi 2011; Monjerezi et al. 2011; Yidana et al. 2011; Ielpo et al. 2012).

Therefore, the present work focuses on the temporal variation and spatial distribution of groundwater contamination using the universal exponential formula based on an immune evolutionary algorithm (IEA-UEF) method, which is intended to prioritize the at-risk areas with high variability potentials of water quality. This will enhance the sustainability of the resource management scheme that is currently far from satisfactory, through a dual water and separate water supply system.

**STUDY AREA AND DATA**

**Geographical situation and climate**

Jilin City is located between 126°25′–126°40′E and 43°46′–44°00′N in Northeastern China, and its urban districts cover an area of about 128 km². The urban area consists of six districts: Jiuzhan, Jiangbei, Hadawan, Centre, Chuangying, and Jiangnan. The site is subject to a temperate continental monsoon climate. The average annual precipitation varies from 650 to 700 mm, and the average annual evaporation is about 1432 mm. The Second Songhua River and its tributaries (Wende River, Mangniu River) flow through the urban area. Figure 1 shows the geographical location of the urban area, as well as the position of the majority of the monitoring wells.

**Geology and hydrogeology**

The study area is located in the Second Songhua River valley plain. The lithology of the aquifer is predominantly strong permeable gravel, circular-gravel, and a pebble layer from the Holocene and upper/lower Pleistocene series. Low mountains and hills around the study area give priority to the bedrock fissured aquifer from the Yanshanian granite (γ2), upper Triassic Dajiagang group (T3d) andesite and the Upper Permian Yangjiagou group (P2y). Because of its poor storage and supply conditions, this aquifer cannot act as a major water source. The utilized aquifer in this study is composed of pore phreatic water in a loose rock mass within the valley plain. Through analysis of the drill-hole
data, it was found that the thickness of the phreatic aquifer is 5-10 m in the valleys of the Wende and Mangniu rivers, and 10-40 m in the valley of the second Songhua River. The water inflow rate per well varies from 100 to 3000 m$^3$/day.

In the region, phreatic water mainly derives from precipitation infiltration, fissure water lateral runoff from the surrounding bedrock and irrigation infiltration. The aquifer discharge consists of lateral runoff from the valley, artificial exploitation, and phreatic water evaporation.

**Groundwater sampling**

Groundwater samples were collected during 1980–2009 from the Water Management office (WMO) of Jilin City, but for unknown reasons, the numbers of sampled wells differed in the WMO from year to year (Table 1). The monitoring aquifer is a Quaternary pore phreatic water layer, and the sample dates are during the dry season (late April to early May). Physical property and chemical analyses of the samples were carried out by the Water Environment Monitoring Center of Jilin province. Eight groundwater quality variables (pH, $\text{SO}_4^{2-}$, $\text{Cl}^-$, $\text{NO}_3^-\text{N}$, $\text{NO}_2^-\text{N}$, $\text{NH}_4^+$-$\text{N}$, $\text{F}^-$, and TH) were studied in this paper, and these reflect the overall characterization of groundwater quality conditions in the study area. Descriptive statistics of the groundwater samples are given in Table 2.

**METHODOLOGY**

**IEA-UEF**

By setting up benchmarks for groundwater quality parameters, a universal exponential formula with a power function suitable for multiple parameters is put forward to evaluate groundwater quality, as first proposed by Li et al. (2008). The IEA is used to optimize formula parameters, and the IEA-UEF is free from the restrictions of types and numbers of pollutants, including the assessment formula of single parameters as a special example. Compared with more traditional methods, this formula is simple, easy to calculate, comparable and practical (Gu et al. 2002; Guo et al. 2006; Ye et al. 2011; Liu et al. 2015). The IEA-UEF equation is described as the following:

$$WQI' = a \left( \sum_{i=1}^{m} W_i x_i \right)^b$$

(1)

where $WQI'$ is the groundwater pollution index, $a$ and $b$ are the parameters to be determined by the IEA, which can establish the range of the $WQI'$ value, $W_i$ is the normalized weight, $m$ is the number of selected groundwater quality variable $i$, and $x_i$ is the specified value of $i$, which can be calculated by Equation (2):

$$x_i = \frac{c_i}{c_{i0}}$$

(2)

where $c_i$ is the value of variable $i$, and $c_{i0}$ is the set benchmark value. The principle of setting $c_{i0}$ ensures that after every $c_i$ ($i = 1, 2, \ldots$) is translated into $x_i$, each specified value will be within one order of magnitude. After repeated trials, 39 reference values of groundwater quality parameters were determined, with $c_{i0}$ listed in Table 3.

**Optimization of parameters of $a$ and $b$**

To make the 39 groundwater quality indicators in Table 3 suitable for the parameters of Equation (1), an optimal

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**Table 1** The number of monitoring wells from 1980 to 2009

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target function should be constructed through Equation (3):

$$
\min F = \frac{1}{Km} \sum_{k=1}^{K} \sum_{i=1}^{m} (WQI_{ik} - WQI_{th})^2
$$  (3)

where $m$ is the number of groundwater quality parameters ($m = 38$ in this paper), and $K$ is the sum of each groundwater sample, when they are optimized. Under the condition of meeting the demands of Equation (3), the parameters $a$ and $b$ are calculated repeatedly by the IEA, in order to optimize these variables. As computed by Matlab, when the optimal value $F$ is less than or equal to 0.0037, the values of $a$ and $b$ are equal to 0.0400 and 0.6472, respectively.

Therefore, the universal formula applicable to the parameters in Table 3 can be described as in Equation (4):

$$
WQI' = 0.0400 \left( \sum_{i=1}^{m} W_i x_i \right)^{0.6472}
$$  (4)

Because the groundwater quality evaluation parameter used to be expressed by real-numbers during the interval of 0 to 100, the $WQI$ should be expanded a hundredfold. The new formula will be as Equation (5):

$$
WQI = 4.00 \left( \sum_{i=1}^{m} W_i x_i \right)^{0.6472}
$$  (5)
As this universal exponential formula was made according to the Quality Standard for Groundwater in China (GB/T 14848-1993), it divides groundwater quality into five classes (see Table 4). This classification only applies to groundwater in general, rather than geothermal water, mine water and brine.

Identification of weight $W_i$

The weight $W_i$ of Equation (5) can be calculated in two cases (Li et al. 2008):

1. When the number of evaluation parameters is large ($m \geq 10$) and the difference between $x_i$ ($i = 1, 2, 3, \ldots, m$) is not significant ($x_{\text{max}}/x_{\text{min}} \leq 10$), every parameter can be seen as equally weighted ($W_i = 1/n$).

2. When the number of evaluation parameters is low ($m < 10$), and $x_{\text{max}}/x_{\text{min}} > 15$, the parameter $W_i$ can be computed as follows:

$$W_i = \begin{cases} \frac{(x_i/2)^{0.5}}{1 - ((1 - x_i/2)^{0.5})}, & 0 \leq x_i \leq 0.5 \\ 1 - ((1 - x_i/2)^{0.5}), & 0.5 < x_i \leq 1.0 \\ 1 + ((x_i - 1)/2)^{0.5}, & x_i > 1.0 \end{cases}$$

(6)

where $x_i'$ is calculated by Equation (7):

$$x_i' = \begin{cases} x_i/100, & x_i \geq 1 \\ 1 - x_i, & x_i < 1 \end{cases}$$

(7)

where $x_i$ is the specified value of the parameter $i$. Finally, $W_i$ can be obtained using Equation (8):

$$W_i = W_i'/\sum_{i=1}^{m} W_i'$$

(8)

RESULTS AND DISCUSSION

Groundwater quality statistics

The groundwater quality analysis revealed that values of pH, SO$_4^{2-}$, Cl$^-$, NO$_3^-$-N, NO$_2^-$-N, NH$_4^+$-N, F$^-$, and TH for the majority of the samples violated Grade III of the Quality...
Standard for Groundwater in China. The minimum levels for this standard are 6.5, 250 mg/L, 250 mg/L, 20 mg/L, 0.2 mg/L, 0.02 mg/L, 1.0 mg/L, and 450 mg/L, respectively. Grade III is the lowest standard under which water can be considered drinkable. Figure 2 shows the averages of eight groundwater quality parameters in six districts during the typical years. Quality varied temporally within districts and spatially between districts where parameters indicated wide ranges and high standard deviations, suggesting that multiple sources and/or complex hydrological processes governed groundwater quality variability (Alaa, 2013). The average pH values had a relatively narrow range of 6.12 to 7.83 (average: 6.75), which is basically similar to neutral water. As Figure 2(a) shows, pH average values that violate the lower limit of 6.5 were reported in six districts between 1985 and 1992. The largest average pH value (7.55) was observed at Chuanying district in 2002, and the lowest value seen at Hadawan district in 1985.

The SO$_4^{2-}$ values ranged from 1.7 to 280.10 mg/L in the entire region, and the average value was 278.40 mg/L. As Figure 2(b) shows, SO$_4^{2-}$ average values of violations of the lower limit of 250 mg/L were reported in six districts between 1985 and 1992. The largest average SO$_4^{2-}$ value (280.10 mg/L) was observed at Chuanying district in 2002, and the lowest value seen at Hadawan district in 1985.

![Figure 2](https://iwaponline.com/ws/article-pdf/16/5/1277/411284/ws016051277.pdf)

**Figure 2** | Temporal variation of the groundwater parameters, (a) pH, (b) SO$_4^{2-}$, (c) Cl$^-$, (d) NO$_3$N, (e) NO$_2$N, (f) NH$_4^+$N, (g) F$^-$, and (h) TH.
shown in Figure 2(b), all values of the typical years are under the red line denoting Grade III of the Quality Standard, which means that none of the averaged groundwater quality parameters violated this classification in the six districts. However, this does not mean that all of the water samples satisfy Grade III of the Quality Standard in the six districts. It also can be seen that the SO$_4^{2-}$ values in 2005 are higher than values in the other typical years. Spatially, the values in Hadawan and Centre districts are generally higher. As time proceeds, the SO$_4^{2-}$ value decreases gradually in Hadawan district. Cl$^-$ showed a large range of between 1.20 and 401.43 mg/L (average: 59.51 mg/L), but the average values of each district comply with the Chinese drinking water standard of 250 mg/L (Figure 2(c)). Similarly, there are some water samples violating the standards of Grade III. Figure 2(c) shows that Cl$^-$ values are relatively stable.

Nitrogen in the form of nitrate (NO$_3^{-}$-N) showed a range of 0.03 to 85.78 mg/L (average: 10.49 mg/L), while nitrite nitrogen (NO$_2^{-}$-N) ranged from 0.00 to 2.39 mg/L (average: 0.08 mg/L). Figures 2(d) and 2(f) show that in almost half of the districts in different typical years, NO$_3^{-}$-N and NO$_2^{-}$-N have exceeded the permissible limits for drinking water (GB/T 14848-1993) of 20.0 mg/L and 0.02 mg/L, respectively, which indicates heavy contamination of the shallow groundwater by nitrate and nitrite. The region of high NO$_3^{-}$-N values is mainly concentrated in most parts of Jiangnan, Chuanying, but also in a small part of Centre. In 2009, the average values (NO$_3^{-}$-N) of the entire region are far below the limit for drinking water (20 mg/L). High NO$_2^{-}$ occurs in almost all of the districts in the early years, but it has decreased with time, especially in 1997 when NO$_2^{-}$ displayed its lowest levels as a whole. High NO$_3^{-}$ concentrations may cause a potentially fatal blood condition known as methemoglobinemia, which affects children younger than 1 year of age (Bokar et al. 2004). In China, high nitrate and nitrite concentrations are known to be major health concerns. High NO$_2^{-}$ concentrations have been revealed to be one of the causes of cancer in China (Lin et al. 2000).

Ammonium as nitrogen (NH$_4^+$-N) was found to fluctuate between 0.00 and 16.87 mg/L, with a mean of 0.60 mg/L. The high average NH$_4^+$ values were distributed mainly within Jiangbei district in different typical years. In 1988, 1992 and 2009, lower average values dominated in most districts. Sources of elevated NO$_3^{-}$, NO$_2^{-}$ and NH$_4^+$ ions in the shallow groundwater of Jilin City included the intensive agricultural activities occurring over the past 60 years, inadequate industrial and domestic waste disposal, and septic tanks systems.

Fluorinion (F$^-$) concentrations showed a small range of 0.00 to 2.37 mg/L (average: 0.27 mg/L). There were minor changes in F$^-$ averages in different years at each district, as shown in Figure 2(g). Among all of the average values of each district during the sampling period, none had exceeded the permissible drinking water limit of 1.00 mg/L. High F$^-$ concentrations has been known to cause human intoxication.

TH displayed a range of 78.05 to 803.13 mg/L (average: 274.43 mg/L). Figure 2(h) shows that only at Jiangnan district in 1980 and 1988, and at Center district in 1980, the TH averages violated the permissible drinking water limit of 450 mg/L, with the hardest water being reported in Jiangnan district in 1988 (average: 560.63 mg/L). Hard water is mainly an aesthetic concern because of the unpleasant taste that a high concentration of Ca$^{2+}$ and other ions give to water. It also reduces the ability of soap to produce lather, and causes scale formation in pipes and on plumbing fixtures. Hard water causes no health risks to water consumers.

**Contamination index mapping**

The water pollution indices in urban areas of Jilin City during 1980 to 2009 have been used for interpolation techniques in drawing groundwater pollution distribution maps, according to IEA-UEF. The Kriging method is utilized for interpolation in this paper. The descriptive graphical representation of the eight typical years with their pollution indices is shown in Figure 3.

From Figure 3, the distribution of groundwater contamination in urban areas of Jilin City in different years could be presented clearly and intuitively. In Figure 3, index values lower than 22.09 were denoted in blue (light blue and dark blue), of which class the groundwater can still be used by the populace. However, index values between 22.09 and 100.00, denoted in yellow and red, show that the groundwater is classified as grades IV and V (Table 4), and the red denotes the worst water quality class.

In general, due to influences from human activity (agricultural, domestic and industrial pollution), the groundwater in much of the study area has been polluted in most years,
which showed high groundwater contamination indices (GCI). In Figure 3, groundwater pollution clearly presents spatial distribution patterns: Jiangbei district showed the highest contamination levels, Center district showed the second-highest contamination levels, and contamination in the other districts was only occasionally present. Temporally, the groundwater contamination level goes through four main stages in the following order: heavy – light – heavy – light. This reflects the transformation of human production and lifestyle from extensive to intensive, from low-technology to high-technology and from manual labour to mechanization.

Jiangbei district is the most important chemical industrial area of Jilin City, so it has experienced the most contamination. As is shown in Figure 3, the GCI in most areas of Jiangbei district are higher than 22.09 from 1980 to 2002, and maximum values reached 100 in some monitoring wells sites. This meant that the groundwater had been heavily polluted and that the water quality belonged to Grades IV and V, as classified using the IEA-UEF. In particular, there are multiple chemical industries, and ash and slag dumps associated with thermal power plants in the northwest part of this district. Hence, the GCI here is generally higher than in the peripheral regions. Since 2002, the dumping of ash and slag had been gradually halted, and urban domestic and industrial sewage discharge was standardized and managed. As a result, the GCI has been decreasing, while the scope of contamination has been diminishing. By 2005 and 2009, only a minor distribution of island pollution appeared in Jiangbei district, which were located in the northwest and southeast areas of the district along the river.

In Center district, periods of heavy contamination mainly occurred in 1980 and 2005, and the scope of pollution was relatively large. In other periods, contamination was infrequent, and the pollution level was low, as shown in Figure 3 whereby the blue region dominated the district (GCI ≤ 22.09). Overall, the GCI decreased gradually from 1980 to 2002. Only in 2005 did the GCI suddenly increase, before decreasing again in 2009. Because Center district, as the old town, has been subjected to pollutants such as industrial waste and urban sewage runoff for many years, the groundwater has been heavily polluted. Until the late
1990s, human living conditions gradually improved, with past seeping wells and pits being filled, and house refuse being transported and cleaned at the same time. As a result, the groundwater pollution intensity weakened, as well as the GCI, in Center district (Figure 3).

In Jiuzhan district, the pollution was mainly concentrated along the Songhua River, and in 1985 and 2002, the groundwater pollution scope and intensity were similar, in which the GCI was greater than 22.09. In other years, the groundwater quality was good and basically stable.

In the southeast of Hadawan district, the groundwater quality was good and stable over several years, which meant that there was little contamination infiltrating the aquifer. The water quality in the northwest part of the district was also good before 1992. Following this, the water quality deteriorated until 2002, during which the maximum GCI was 43.07 in the region in 2002. Subsequently, the GCI has been decreasing. It is known that Hadawan district, as an important vegetable production base, was subject to pollutants derived from chemical fertilizers and pesticides over many years, with groundwater pollution intensity reaching a peak in 2002. Then, with further urbanization, the area of cultivated land gradually decreased. Since 2002, the groundwater quality has also been visibly decreasing, in association with these land use changes.

Chuanying district used to be on the outskirts of Jilin City, but since the 1980s it has gradually been developing into an urban area. With increases in human activities, the GCI has also increased slowly. The groundwater pollution level improved after 2005, and in 2009, the average GCI approached 10, which is characterized as Grade II.

In the early period, as Jiangnan district had not been developed and utilized, the groundwater quality there was good. Subsequently, Jiangnan district was transformed into a vegetable production base and the groundwater quality became worse, with a maximum GCI of 85.32 being observed in 1988. Since 1990, Jiangnan district has changed into a high-technology development region. Under disturbances due to intensive human activities, the GCI here increased gradually, as did the scope of pollution during the early stage. Afterwards, with technological development and productive technology improvements, the discharge of sewage has diminished and the quality of wastewater has improved. The groundwater environment became much ameliorated, and this district is now viewed as an ecologically friendly and habitable new city.

CONCLUSIONS

This study reports the first findings concerning the spatio-temporal status of shallow groundwater quality in urban areas of Jilin City from 1980 to 2009, using the IEA-UEF method. In the current study, the following conclusions were reached:

1. The IEA-UEF is a good groundwater contamination index model, which can reflect the status of groundwater pollution clearly and intuitively. Meanwhile, the partition interpolation depends on the boundary of the Songhua River using the Kriging method, which solves the discontinuity problem of groundwater pollution.

2. NO₃⁻, NO₂⁻, NH₄⁺, and TH were the primary parameters that influenced groundwater quality, and most values violated Grade III of the Chinese Quality Standard for Groundwater in a large proportion of groundwater samples.

3. The ever-increasing population, unplanned city growth, excessive use of fertilizers and pesticides in agricultural land, lack of proper sewage systems, and improper disposal of wastewater (from both household and industrial activities) collectively led to deterioration of the groundwater environment in Jilin City.

4. Spatially, Jiangbei district was more heavily polluted than the other districts. The contamination distribution is consistent with proximity to contamination sources, topography, and urban, agricultural, and industrial practices. Temporally, the most significant level and the most extensive scope of groundwater contamination were both observed in 1985 and 2002. The groundwater contamination status in the study area generally underwent four stages during 1980 to 2009: heavy – light – heavy – light.

This research line is also not concluded yet. In future research, we would collect and analyse more data in order to identify whether the drinkability condition (Grade III) is being systematically violated or it is due to specific days/months/out layers instead. In addition, we would like to find out whether the drinkability condition is being systematically violated or it is due to specific days/months/out layers instead.
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