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GROUNDWATER QUALITY AND CONTAMINATION INDEX MAPPING IN CHANGCHUN CITY, CHINA

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ABSTRACT: Groundwater in Changchun City, Jilin Province of China tends to be influenced by human activities. Chemical types of groundwater were detected in both shallow and deep groundwater were: HCO_3^- - Ca^{2+} and HCO_3^- - Ca^{2+}•Mg^{2+} or HCO_3^- - Mg^{2+}•Ca^{2+}; SO_4^{2-} - Ca^{2+} and SO_4^{2-} - Ca^{2+}•Mg^{2+}; Cl^- - Ca^{2+}; and CO_3^{2-} - Na^+. The deteriorations of groundwater quality due to the increase of TDS, NO_3^- + NO_2^- (as Nitrogen) and TH contents have been observed from 1991 to 1998. Scatter analyses showed strong positive correlations between Ca^{2+}, Cl^- and NO_3^- ions and weak negative correlations between the depth of water table and Ca^{2+}, SO_4^{2-}, Cl^- and NO_3^- ions. A mapping of contaminant index based on Chinese standard of groundwater showed that a large proportion of the groundwater in 1998 was deteriorated by human process. Despite their low values of sodium adsorption ratio (SAR), the most of the sampled wells were not suitable for drinking and agriculture purposes due to higher contents of NO_3^-, NO_2^- and Mn^{2+} ions.
KEY WORDS: hydrochemistry; groundwater quality; contaminant index; linear regression analysis; GIS mapping; Changchun City

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1 INTRODUCTION

Groundwater in Changchun City assures about 45% of total water supply. Drinking water supply of Changchun City was mostly served by surface water from Shitoukoumen and Xinlicheng reservoirs located in the east and south of the city (ZHANG, 1993). However, with the development of urban construction the groundwater especially from deep boreholes is also used for drinking purpose in suburban areas. The urbanization process in Changchun City threatens the groundwater quality and already there have been increasing cases reported on the groundwater deterioration especially the shallow groundwater due to high total hardness, high intensity of mineralization and high content of Cl^-, SO_4^{2-} and NO_3^- (ZHANG, 1993).

The aim of this study is to investigate and characterize dynamic changes in the groundwater quality in Changchun City over the past years. Earlier studies have noted some of the changes (ZHAO, 1990; MOU and CHENG, 1992; ZHANG, 1993; ZHU, 2001), however there are few investigative demonstrations to their causes. From the results presented here, it is evident that portions of major ions (Ca^{2+}, Cl^-, SO_4^{2-} and NO_3^-) are of human origin. The results of this study will contribute greatly to a better understanding for the hydrochemistry and the evolution of water quality in Changchun City.

2 STUDY AREA

The study area is located in the urban area of Changchun City (Fig. 1). The city, capital of Jilin Province, is situated in the central part of Northeast China Plain at 43°47′ to 43°59′ N and 125°12′ to 125°26′ E. It covers a total area of about 500km^2. Its climate is generally characterized by cold and long winter (November to February), and short summer (June to August) seasons. The mean annual precipitation in Changchun City is in the range of 500–600mm and mean annual temperature is 4℃.

The population of Changchun City is about 2×10^6. The city has witnessed a rapid urbanization and economic growth over the last ten years. This has lead to rapid increase in population, industries and consequently a deficit in water supply. In addition many environ-

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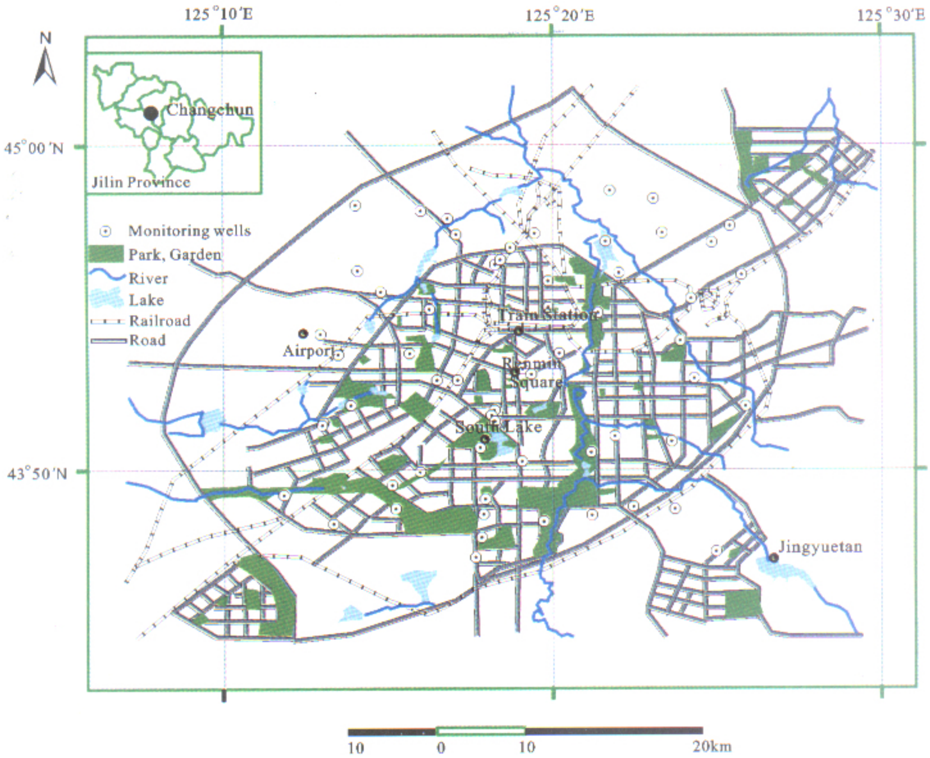

Fig. 1 Description of the study area

mental problems such as water pollution, solid wastes and sewage effluents management have been increased. Although there are several artificial lakes, reservoirs for water supply and wastewater drains in the city, the Yitong River is the main surface water body in the study area.

The geological structure of the study area is constituted of the Quaternary deposits in the upper part and the Cretaceous and the Jurassic bedrock in the lower part. The Quaternary formations, formed by alluvium from the river, constitute of backfill soil, clay soil and sandy soil with thickness varying from 3–20m. (Fig. 2). The Cretaceous formations are mostly constituted of mudstone and argillaceous sandstone with about 300m of an average thickness. The Jurassic bedrock is constituted of rhyolite and andesite. The tectonic is characterized by vertical and horizontal displacements in the Cretaceous and the Jurassic formations (Fig. 2). The major faults responsible for these structures are generally hidden by the Quaternary formations.

Two main types characterize the aquifer systems. The unconsolidated Quaternary formations constitute the upper aquifers, which are exploited through shallow wells. Their thickness range from 5m to 20m while productivity varies from $10m^3/d$ to $1000m^3/d$.

Deep-fissured aquifers are the result of fractured and fissured Cretaceous and Jurassic formations. Fractured sandstone is a very productive aquifer while the less fractured mudstone is a very poor one. Their thickness range from 20m to 300m while productivity varies from $100m^3/d$ to $3000m^3/d$.

3 MATERIALS AND METHODS

The Water Resources Office of Changchun City and the College of Environment and Resources of Jilin University have conducted the groundwater quality monitoring programs since 1981 in Changchun City. The program, based on annual sampling of randomly selected municipal water supply wells from July to September, was designed to provide data for characterization of the groundwater quality in the shallow Quaternary and deep bedrock aquifers. The water temperature and pH were measured in the field. The concentrations of Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, NO_3^- and SO_4^{2-} ions were determined by titration while those of K^+, Na^+ and Fe^{3+}, Mn^{3+} were determined by atomic absorption spectrometry.

In order to find the source of the pollution, regression analyses were used to determine relationships between nitrate and depth of water (thickness of the unsaturated zone) and with other ions. A mapping using GIS features was done to determine the zones that are mostly affected by inorganic pollution according to the Chinese quality standard for groundwater (GB/T 14848-1993).

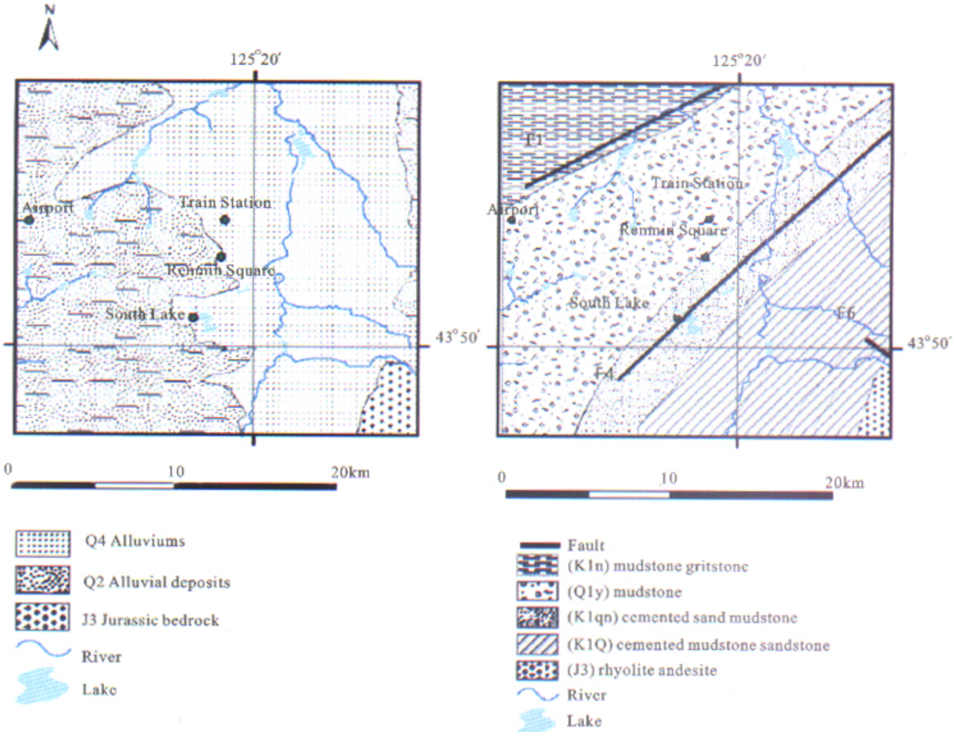

Fig. 2 Geological map of the aquifer system (left shallow aquifer, right deep aquifer)

4 RESULTS AND DISCUSSION

4.1 Groundwater Quality

Table 1 shows the statistics of chemical analyses in shallow and deep samples wells in 1991 and 1998. The maximum and mean values of most of the groundwater constituents were higher in shallow wells compared to those of deep wells during the two monitoring years.

The chemical types of water were classified according to the three major cations and three major anions. The cations are calcium (Ca^{2+}), magnesium (Mg^{2+}) and sodium (Na^{+}), while the anions are bicarbonate (HCO_3^-), chloride (Cl^-) and sulfate (SO_4^{2-}). The concentration of potassium is usually grouped with sodium in the determination of water chemical type. The data from 46 and 59 water samples collected in 1991 and 1998 respectively, for both shallow and deep groundwater, were analyzed using Piper diagram (PIPER, 1940) shown in Fig. 3.

The following deductions can be made based on the two Piper diagrams (Fig. 3). There were 4 classes of water chemical types for both shallow and deep groundwater: 1) HCO_3^- - Ca^{2+} and HCO_3^- - Ca^{2+}•Mg^{2+} or HCO_3^- - Mg^{2+}•Ca^{2+}; 2) SO_4^{2-} - Ca^{2+} and SO_4^{2-} - Ca^{2+} • Mg^{2+}; 3) Cl^- - Ca^{2+}; and 4) CO_3^{2-} - Na^+. In 45 % of the water samples in 1991 and 1998 there was a dominance type of HCO_3^- •CO_3^{2-} over the Cl^- •SO_4^{2-} type while the other 55% indicated the opposite case. Shallow groundwater was mostly classified into the following chemical types: SO_4^{2-} - Ca^{2+} and SO_4^{2-} - Ca^{2+}•Mg^{2+}; or Cl^- - Ca^{2+} while deep groundwater was classified into HCO_3^- - Ca^{2+} and HCO_3^- - Ca^{2+}•Mg^{2+}. There are very few samples for CO_3^{2-} - Na^+. The Ca^{2+}, Mg^{2+}, HCO_3^-, Cl^-, and SO_4^{2+} ions are generally found at higher concentrations by compared to the other major ions (Na^+, K^+ and CO_3^{2-}). The pH ranged from 7 to 8 for both shallow and deep groundwater, which showed a neutral to weak alkaline water.

The water temperature values ranged from 8℃ in the shallow aquifers up to 40℃ in the deep aquifers. In 1991 and 1998 the total hardness ranged from 13mg/L to 1000mg/L and 7mg/L to 954mg/L $CaCO_3$ for shallow and deep monitoring wells respectively.

In order to visualize the distribution and the evolution of groundwater parameters over the time boxplots were used. The boxplots are efficient method to visualize how individual points are distributed in datasets. The box itself outlines the range of half the data (the 25th to the 75th percentiles). The bar inside the box showed the median, or the 50th percentile. Two whiskers (vertical lines extending from the box) indicated the distance to the highest and lowest data points, which were not outliers. Outliers indicated by points above or below the whiskers. If the median was located midway between top and bottom of the box, the data were normally distributed.

Table 1 Range of chemical parameters in groundwater of Changchun City in 1991 and 1998

Ground water	Parameter	N	Min	Max	Mean	Std dev	Ground water	N	Min	Max	Mean	Std dev
Shallow water 1991	pH	35	7.23	8.58	7.56	0.99	Deep water 1991	12	7.44	8.48	8.01	0.27
	TDS (g/L)	35	0.10	1.70	0.71	0.45		12	0.15	1.70	0.48	0.42
	Ca^{2+} (mg/L)	35	5.99	351.72	113.46	86.94		12	2.00	285.70	60.00	69.00
	Cl^- (mg/L)	35	5.73	292.85	106.94	84.08		12	5.75	255.00	52.85	70.70
	TH (mg/L $CaCO_3$)	35	20.05	1000.00	377.00	273.00		12	7.54	957.00	205.00	232.00
	HCO_3^- (mg/L)	35	40.60	319.00	165.82	85.26		12	45.57	320.00	170.88	75.82
	K^+ (mg/L)	35	0.30	46.00	3.26	8.06		12	0.50	4.10	1.07	1.04
	Mg^{2+} (mg/L)	35	1.20	65.70	22.51	18.64		12	0.61	58.00	13.42	15.54
	Na^+ (mg/L)	35	11.00	140.00	51.36	36.43		12	9.20	267.50	51.58	68.83
	SO_4^{2-} (mg/L)	35	6.00	460.00	93.45	98.45		12	12.00	460.00	71.57	126.54
	F^- (mg/L)	35	0.00	0.70	0.15	0.16		12	0.06	1.04	0.51	0.28
	NH_4^+ (mg/L)	35	0.00	13.00	0.51	2.34		12	0.00	0.30	0.03	0.08
	NO_3^- (mg/L)	35	0.00	580.00	126.47	159.31		12	0.00	230.00	32.99	60.68
	NO_2^- (mg/L)	35	0.00	3.76	0.31	0.73		12	0.00	0.30	0.02	0.05
	Total Fe (mg/L)	35	0.00	24.00	2.55	6.11		12	0.00	1.00	0.23	0.37
Shallow water 1998	TDS (g/L)	38	0.29	1.21	0.67	0.26	Deep water 1998	21	0.19	0.91	0.50	0.20
	Ca^{2+} (mg/L)	38	19.00	305.00	112.99	66.10		21	12.10	178.40	66.83	45.30
	Cl^- (mg/L)	38	21.00	363.00	113.28	86.31		21	0.00	151.00	49.23	44.07
	TH (mg/L $CaCO_3$)	38	13.00	758.00	199.14	179.00		21	79.97	522.00	214.00	128.32
	HCO_3^- (mg/L)	38	20.00	428.00	185.24	91.88		21	92.30	444.30	225.56	91.51
	K^+ (mg/L)	38	0.00	204.00	6.52	32.95		21	0.30	1.10	0.50	0.20
	Mg^{2+} (mg/L)	38	1.00	182.00	21.98	28.37		21	0.00	34.78	11.48	8.48
	Na^+ (mg/L)	38	17.00	389.00	48.00	60.47		21	11.00	133.00	45.62	34.18
	SO_4^{2-} (mg/L)	38	0.00	340.00	86.27	64.65		21	2.00	99.40	43.91	23.16
	F^- (mg/L)	38	0.00	6.00	0.44	0.90		21	0.20	0.66	0.40	0.14
	NH_4^+ (mg/L)	38	0.00	3.00	0.39	0.56		21	0.00	9.00	0.65	1.92
	NO_3^- (mg/L)	38	0.00	364.00	83.98	77.84		21	0.00	220.00	39.52	52.43
	NO_2^- (mg/L)	38	0.00	15.00	4.45	4.00		21	0.00	10.01	0.55	2.17
	Total Fe (mg/L)	38	0.04	24.00	1.80	4.27		21	0.00	4.30	0.90	1.06
	Mn^{2+} (mg/L)	38	0.00	4.40	0.42	0.85		21	0.00	3.00	0.37	0.92

Note: N is number of well.

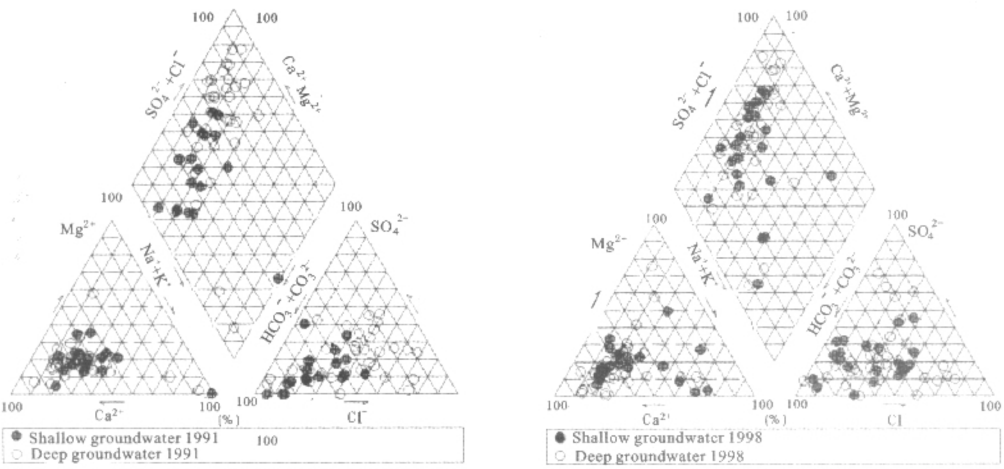

Fig. 3 Piper diagram showing the chemical composition of groundwater in Changchun City in 1991 and 1998

Boxplots of groundwater quality parameter: TDS, nitrogen (NO_3^- +NO_2^- as N), Total Fe, and TH of the shallow and the deep wells data are shown in Fig.4. The Total Dissolved Solids (TDS) is the amount of solids left when a filtered ground water sample is evaporated to dryness. TDS can also be calculated from major ion concentrations. The MCL (Maximum Contaminant Level) for TDS is 500mg/L. From 1991 to 1998 the TDS concentrations (calculated from major ion concentrations) of the shallow groundwater ranged from 0.11g/L to 1.8g/L with the median varying from 0.4g/L to 0.7g/L. The data are mostly normally distributed. The medians exceeded the MCL 500mg/L for drinking water in 1991, 1992, 1993, 1994, 1997, 1998,

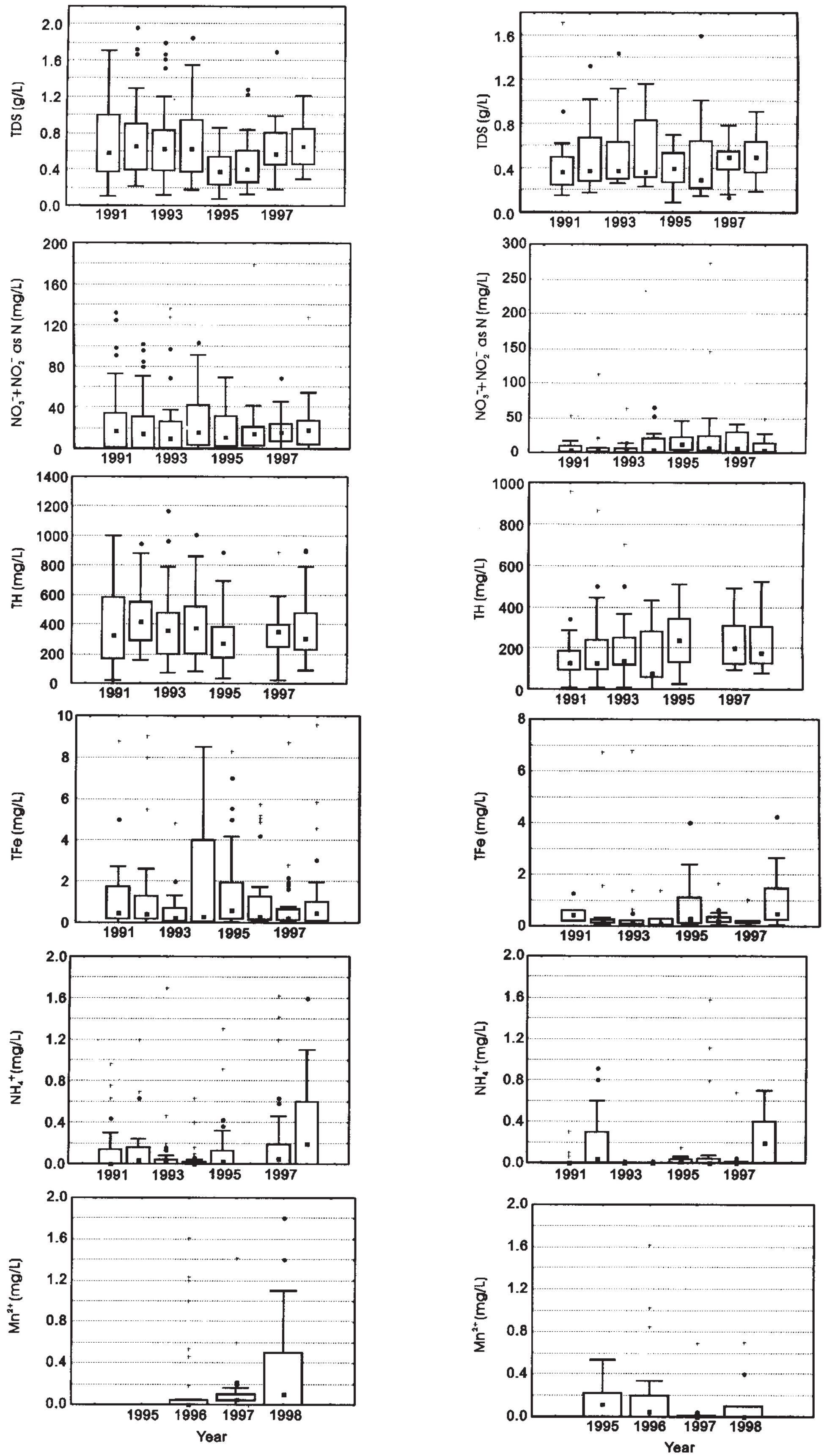

Fig. 4 Boxplots showing the distribution and the evolution of the TDS, $NO_3^-+NO_2^-$ (as N), TH, TFe, NH_4^+ and Mn^{2+} concentrations data of the shallow (left) and deep (right) groundwater of Changchun City over the years

which show high mineralization. At the sites where TDS exceeded 1000mg/L, Ca^{2+}, SO_4^{2-}, Mg^{2+}, Cl^-, and NO_3^- concentrations were elevated while Na^+ concentrations were higher in the sites with lower TDS values. TDS data of the deep groundwater from 1991 to 1998 ranged from 0.1g/L to 1.3g/L lower than those of the shallow groundwater, the median values varied from 0.3mg/L to 0.5mg/L. The data showed strong asymmetric distributions. For both shallow and deep groundwater the TDS median values showed an upward trend, which explains a deterioration of groundwater quality over the time.

The $NO_3^- + NO_2^-$ (as nitrogen) levels in the shallow groundwater ranged from below 0.05mg/L of the limit in the laboratory to higher values of over 100mg/L. The median values varied from 10mg/L (the recommended level for drinking water) to 20mg/L, which indicates a serious contamination of the shallow groundwater by nitrate. The data were normally distributed and showed a slight increase of median values over the time. The $NO_3^- + NO_2^-$ slightly influenced the deep groundwater, but the data were non-normally distributed. The non-outlier values ranged from 0.5mg/L to 50mg/L with median values varying from 0.5 mg/L to 10mg/L. High NO_3^- concentrations may cause a potential-fatal blood condition known as methemoglobinemia, which affects children lower than 1 year old. In China high nitrate and nitrite concentrations are known as major health concern. High NO_2^- concentrations have been revealed to be one of the causes of cancer diseases in China (LIN *et al*., 2000).

The ammonium (NH_4^+) concentrations for the shallow groundwater of Changchun City from 1991 to 1998 ranged from 0mg/L (less than the detection limit of 0.01mg/L) to 2mg/L with the median value being 0.1mg/L to 0.3mg/L. The distribution was skewed due to high values at some sites, the higher values could be indicative of fertilizer sources. The NH_4^+ values in the deep ground water were lower than that in the shallow groundwater. Specifically sources of elevated NO_3^-, NO_2^- and NH_4^+ ions in the groundwater of Changchun City include the intensive agricultural activities over the past 50 years, inadequate waste disposal, and septic tanks systems and also a poor water well construction especially in the shallow aquifer.

From 1991 to 1998, 50% of the monitoring wells for both the shallow ground and the deep groundwater had dissolved higher irons and more than 0.3mg/l of recommended level. The distribution data of total Fe ions was skewed due to high values at some sites. The median values exceed 0.3mg/L of the recommended level. Available data of Mn^{2+} ions were monitored from 1995 to 1998. The median values were varying from 0.05 mg/L to 0.2 mg/L for both shallow and deep groundwater. Due to high concentration levels of Mn^{2+} ions, 39% of shallow wells and 19% of deep wells were not suitable for drinking purpose in 1998. Mn^{2+} concentration levels being higher than 0.05mg/L can have adverse affects on health especially the nervous system.

Hardness was calculated from the Ca^{2+} and Mg^{2+} concentrations in the water samples. From 1991 to 1998 the total hardness values in the shallow groundwater of Changchun City ranged from 20mg/L to 1000 mg/L (as $CaCO_3$) with the median value varying from 300mg/L to 400mg/L. Except for those of 1997 and 1998 the data were normally distributed (The data of Ca^{2+} ions concentration in 1996 are not available). The median values were higher than 150mg/L of the recommended value. The non-outlier values of TH data of the deep groundwater from 1991 to 1998 ranged from 7mg/L to 500mg/L with median varying from 100mg/L to 200mg/L. The datasets were not well distributed. There is an increase of the total hardness values over the time. Moreover, fluorides concentrations remained low from 1991 to 1998 the mean values ranged from 0.1mg/L to 0.5mg/L, in contrast to many parts in China, where high fluoride concentrations in groundwater was a major environmental problem (LIN *et al*., 2000).

According to Chinese standard of groundwater (GB/T 14848-1993), in 1998, of the total 38 shallow monitoring wells, only one well (A0573) and 3 wells (A0573, B018, A035) were suitable for drinking water and agriculture purpose respectively. Out of the 21deep wells, 3 wells (A107, A062, and A092) and 5 wells (A0162, A0107, A062, A092, and B014) were suitable for drinking and agriculture purpose respectively while there was no significant cation exchange between Ca^{2+} and Na^{2+}, and the values of sodium adsorption ratio (SAR) are very low with only 3 among the 59 samples in 1998 presenting a SAR greater than 10.

4.2 Statistical Analysis

Scatter analyses based on parametric Pearson correlation indicates significant positive correlations between NO_3^- and Ca^{2+} and Cl^- in 1991, 1995 and 1998 in both shallow and deep groundwater (Table 2).

Weak negative correlations existed in 1998 between the depth of water (thickness of the unsaturated zone) and Ca^{2+}, SO_4^{2-}, NO_3^- ions in deep groundwater. The strong correlation between NO_3^-, Ca^{2+} and Cl^- explains the human origin of part of the Ca^{2+} and Cl^- ions.

The weak negative correlations between the depth of

Table 2 Correlation relationships between parameters in the shallow and deep groundwater

Parameters	Pearson correlation coefficient					
	Shallow well in 1991	Deep well in 1991	Shallow well in 1995	Deep well in 1995	Shallow well in 1998	Deep well in 1998
Ca^{2+} and NO_3^-	0.861[2]	0.967[2]	0.572[2]	0.703[2]	0.756[2]	0.651[2]
Ca^{2+} and Cl^-	0.885[2]	0.956[2]	0.797[2]	0.780[2]	0.833[2]	0.766[2]
Cl^- and NO_3^-	0.729[2]	0.903[2]	0.573[2]	0.691[2]	0.558[2]	0.761[2]
Water table depth and Ca^{2+}					0.039	−0.233
Water table depth and SO_4^{2-}					0.075	−0.204
Water table depth and NO_3^-					0.104	−0.252
Water table depth and Cl^-					0.014	−0.539[1]
Number of well (*N*)	35	15	55	24	38	21

Notes:1. Correlation is significant at the 0.05 levels; 2.Correlation is very significant at the 0.01 levels

water table and some major ions (Ca^{2+}, Cl^-, SO_4^{2+}, and NO_3^-) confirm the fact that portions of these ions are loaded from the unsaturated zone.

4.3 Contamination Index Mapping

The main water pollutants in the study area monitored in 1998 such as NO_3^-, NO_2^-, NH_4^+, Mn^{2+}, and total Fe have been used for the mapping. The mapping was done according to Chinese quality standard for groundwater (GB/T 14848-1993).

Table 3 showed the classification of groundwater according to concentrations of the mains pollutants in the study area. Some values are assigned according to the grade.

Table 4 showed the repartition of the assigned values according to grade.

Table 3 Classification of water quality according to concentration of pollutants

Ions (mg/L)	Standard class values				
	I	II	III	IV	V
Total Fe	≤ 0.1	≤ 0.2	≤ 0.3	≤ 1.5	>1.5
Mn^{2+}	≤ 0.05	≤ 0.05	≤ 0.1	≤ 1	>1
NO_3^-	≤ 2	≤ 5	≤ 20	≤ 30	>30
NO_2^-	≤ 0.00	≤ 0.1	≤ 1	≤ 2	>2
NH_4^+	≤ 0.02	≤ 0.02	≤ 0.2	≤ 0.5	>0.5

Table 4 Repartition of given values according to grade (GB/T 14848-1993)

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

The contamination index was then computed using the formula:

$$F=\sqrt{\frac{\overline{F}^2+F_{max}^2}{2}} \quad (1)$$

where

$$\overline{F}=\frac{1}{n}\sum_{i=1}^{n}F_i \quad (2)$$

F_i is a given value for the pollutant, i is the grade (Table 3). $F_{max.}$ is the highest F_i value for a pollutant i. n is number of pollutant.

A nomenclature was given to the water quality according to the contamination index F (Table 5):

Table 5 Nomenclature according to index value (GB/T 14848-1993)

Water quality	Very Good	Good	Moderate	Bad	Worst
F	<0.80	0.80–2.50	2.50–4.25	4.25–7.20	>7.20

GIS features were used for the mapping. ModelBuilder, an extension of ArcView spatial analysis (ESRI, 1998) and ArcView GIS statistical analysis (LEE, 2001) were used for the mapping process. A grid concentration map for each pollutant was generated by Kriging method with an error < 5% and reclassified according to Table 3. The contamination index maps for both shallow and deep groundwater (Fig. 5) were then the results of arithmetic overlay of all concentration maps by computing F through ModelBuilder. Index values lower than 0.8 were almost nonexistent. Fifty percent of the shallow aquifer was occupied by the worst water quality (contamination index greater than 7.2) while 80% of the deep aquifer was occupied by the bad water quality index (contamination index between 4.25 and 7.2).

4 CONCLUSIONS

Four classes of water chemical types for both shallow and deep groundwater of Changchun are HCO_3^- - Ca^{2+} and HCO_3^- - Ca^{2+}•Mg^{2+} or HCO_3^- - Mg^{2+}•Ca^{2+}; SO_4^{2-} - Ca^{2+} and SO_4^{2-} - Ca^{2+}•Mg^{2+}; Cl^- - Ca^{2+}; and CO_3^{2-} - Na^+. High NO_3^- + NO_2^- (as nitrogen), Mn^{2+} and total dissolved Fe ions content combined with higher TDS and TH values were observed in the study area. Their concentration levels and distributions are an issue of concern to Changchun City.

In 1991, 1995 and 1998; NO_3^- ions were significantly positively correlated for the shallow and deep ground water, especially with major ions such as Cl^- and Ca^{2+}, which indicates a human origin of these ions. The weak negative correlations were shown between the depths of

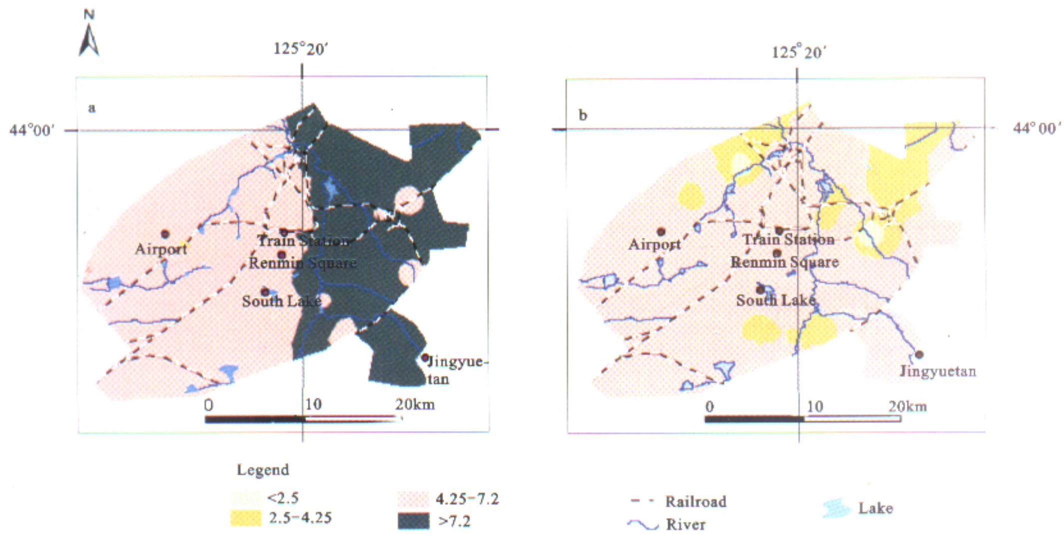

Fig. 5 Contamination index maps of shallow(a) and deep(b) groundwater in Changchun City in 1998

water table and Ca^{2+}, NO_3^-, SO_4^{2-} and Cl^- ions in the deep groundwater, explaining the influence of fractures and fissure on the process of contamination in the deep fractured aquifers of Changchun City. These trends recorded by this study highlight the need for long-term studies to understand about the hydro-chemical and human processes. An environmental isotope study is also necessary to confirm these hypotheses. The contaminated surfaces generated by the mapping process showed that half portion of the city′s shallow groundwater in 1998 was located in the worst water quality index area (contamination index was higher than 7.2), while 80% of the city′s deep groundwater was located in the bad water quality indexed area (contamination index was between 4.25 and 7.2).

This study, which is based on a monitoring of a large number of chemical species, would be useful for environmentalist and decision-makers.

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