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Study of the mechanism that increases the hardness of groundwater: A case study from Beijing urban area

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The rapid development of construction and the increase of population water demand were increased rapidly in Beijing, China's capital city, yet one of its fastest-growing municipalities is running out of water. Today, more than 70% of the municipality's total water supply comes from groundwater. Therefore groundwater has been over-exploited a long time which led to regional water level down and more and more groundwater is becoming polluted, which has disadvantage on drinking water. As a consequence of a series of physico-chemical and biochemical reactions, including dissolution, ion-exchange and reactions involving organic compounds, the calcium and magnesium in the infiltrating solutions have increased. The solutions have infiltrated into aquifers and increased the hardness of the groundwater; over-abstraction of groundwater has accelerated the process. The ISD = f (ISC) diagram indicates that central part of Beijing's groundwater is relatively recent with a speed of mean circulation.

Key words: Development, water demand, pollution, groundwater, hardness, nitrate.

INTRODUCTION

All life forms on the earth contain water and water is crucial for any life form on the earth. Apart from being the essential ingredient of living organisms, water has numerous other uses and benefits. Groundwaters form a circle of the natural hydrologic chain like surface waters and the other water in the atmosphere. Hydrologic, hydraulic and geologic processes play important roles during underground water's formation, storage, underground flow and coming up to the surface of the earth (Mande et al., 2011).

As recently as 30 years ago, Beijing residents regarded groundwater as an inexhaustible resource (Tang Ninghua and Huan, 1973). Now hydrogeologists warn that the groundwater is running out. With the development of urban's construction, industry, agriculture, and the increase of the population and economy, demands for water-resource grow rapidly. Therefore, Beijing's groundwater table is dropping, water is being pumped out faster than it can be replenished, and more and more groundwater is becoming polluted. Today, more than 70% of the municipality's total water supply comes from groundwater (Wang and Yuan, 1981; Tang Ninghua and Huan, 1973). The rest is surface water coming from Beijing's dwindling reservoirs and rivers.

The hardness of water is due to the presence of polyvalent metallic ions, principally Ca^{2+} and Mg^{2+} (Crittenden and Montgomery Watson Harza (Firm) 2005). There are many negative aspects resulting from hard water both for domestic and industrial usage. For example, in lather production, if the water is hard, it

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Table 1. Degree of hardness as a function of the hardness dosage.

Hardness, mg/L as $CaCO_3$	Degree of hardness
1-75	Soft
75-150	Moderately hard
150-300	Hard
300 and more	Very hard

requires considerable amounts of soap before a lather can be produced. In the ordinary life, hard water produces scale in hot water pipes, heater, boilers, and other units where the temperature of the water is increased appreciably. The chemical equation for this process is showing below:

$$Ca^{2+} + 2HCO_3^- \rightarrow CaCO_3 + CO_2 + H_2O \qquad (1)$$

Where $CaCO_3$ is the main component for the scale when hard water is heated (Reynolds and Richards, 1996).

Hardness of waters varies considerably from place to place. In general, groundwaters are harder than surface waters. Hardness can be expected in regions where large amounts of limestone are found, since water with carbon dioxide will dissolve limestone, releasing the calcium ion. Hardness is measured in terms of milli-equivalents per liter or equivalent $CaCO_3$, and the degree of hardness was listed in many books, Table 1 (Reynolds and Richards, 1996).

Knowing the hardness of water is important in evaluating its use as a domestic or industrial water supply. The hardness must be known in determining the amount of chemicals required for lime-soda softening and in the design of ion exchange softening units.

On the other hand, public acceptance of hardness varies from community to community, consumer sensitivity being related to the degree to which the consumer is accustomed. Because of this variation in consumer acceptance, finished water hardness produced by different utility softening plants will range from 50 mg/L to 150 mg/L as $CaCO_3$. Based on the Table 1, the hardness range of finished water covers the scale from soft water to hard water (Letterman and American Water Works Association, 1999).

In this study, groundwater hardness quality at central Beijing urban area was carried out. A comprehensive investigation of the natural and artificial factors that increase the hardness of groundwater was made to establish a clear relationship between hardness and groundwater quality parameters.

Study area

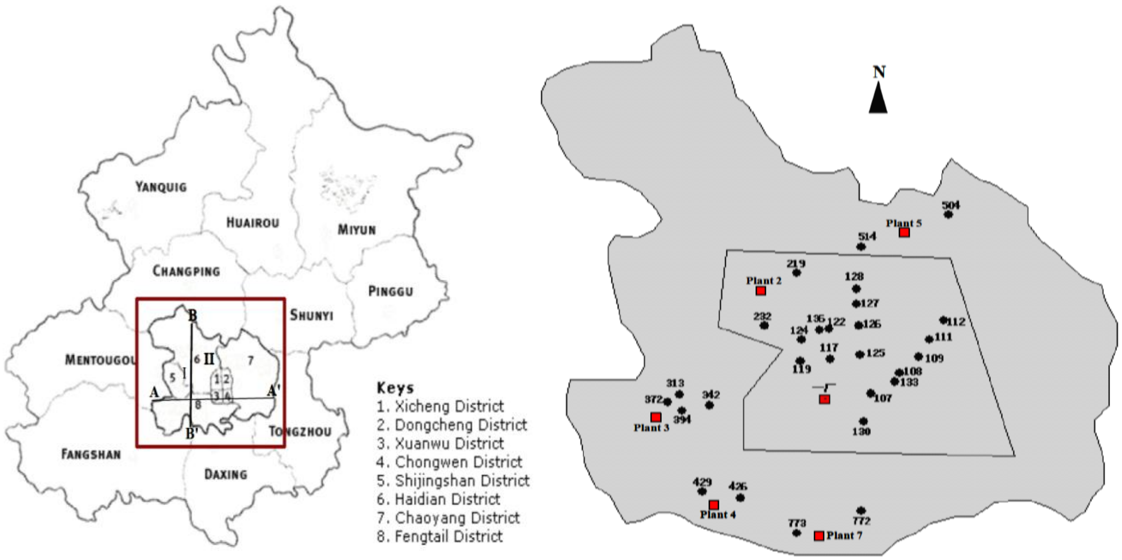
The study area located between latitudes 39°49'51.52"-40°00'38.01"N and longitudes 116°14'45.70 - 116°31'14.19"E covers an area of 1300 km^2 (Figure 1).

The study area includes urban core (Xicheng District [1], Dongcheng District [2], Xuanwu District [3] and Chongwen District [4]) and Inner Suburbs (Shijingshan District [5], Haidian District[6] Chaoyang District [7], and Fengtai District [8]) were once considered on the city's outskirts, but are now integral parts of the city inside the 5th Ring Road. The climate is a monsoon-influenced humid continental climate, characterized by hot, humid summers due to the East Asian monsoon, and generally cold, windy, dry winters that reflect the influence of the vast Siberian anticyclone. The average temperature is about 11.7°C with the highest temperature recorded was 42°C and the lowest recorded was -27°C. Most of the area is flat plain, where the surface elevation ranges between 59 m to 33 m a.s.l.

Geology and hydrogeology

The area discussed below ranges from eastern xishan mountains of Beijing to upper and middle part of Yongding River cover an area about 1300 km^2. The main aquifers of the central of Beijing are the alluvial gravel-sand sediments of the Quaternary period. The line (B-B') from Haidian to Fengtai divides this region into two parts: the unconfined aquifer consisting of a single gravel stratum in the west and the confined aquifer consisting of multiple gravel and sand strata in the east (Figure 2). The plain slopes in a south easterly direction. The Yongding and Chaobai rivers run through the whole area. These are an important source of recharge for the groundwater used in Beijing City. Beijing's rainfall varies geographically, seasonally and annually. Eighty-five percent of the annual precipitation falls between July and September. Rainfall also varies between the sub-watersheds within the municipality and particularly between mountainous areas and the low-lying plain. Beijing's average yearly precipitation is 590 mm with a recorded high of 1,406 mm in 1959 and a low of 242 mm in 1869. Beijing has recorded 25 years of drought since the 1970s. Drought, in this context, is broadly defined as below-average rainfall. Meteorologists in Beijing define an extremely dry year as 300 mm of rainfall or less, which is about 50% below average. A moderately dry year is defined as 450 mm of rainfall or about 25% below average. Between 1999 and 2008, Beijing's average annual precipitation was 428 mm or 28% below average. Figure 3 summarizes the decline in average annual precipitation.

The shallow groundwater on the Beijing plain is recharged 44% by precipitation and 31% by seepage of surface water. As the area covered by buildings and roads has increased dramatically in recent years, less water is able to seep through the ground naturally. Both the rate and volume of groundwater recharge are decreasing. Continued fall in groundwater level, persistent drought climate and insufficient surface water, will greatly threaten the water supply in the Beijing central



I area of unconfined aquifer

II area of confined aquifer

Figure 1. Plan of Beijing City and suburban area showing the distribution of observation wells.

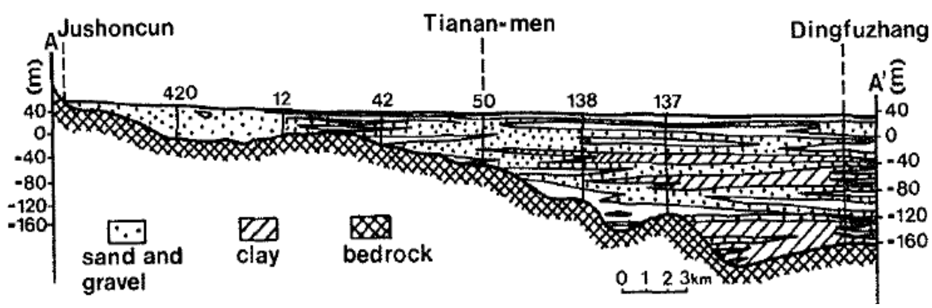


Figure 2. Geological sketch showing Quaternary aquifers in the Beijing area (A-A' geological section: Figure 1).

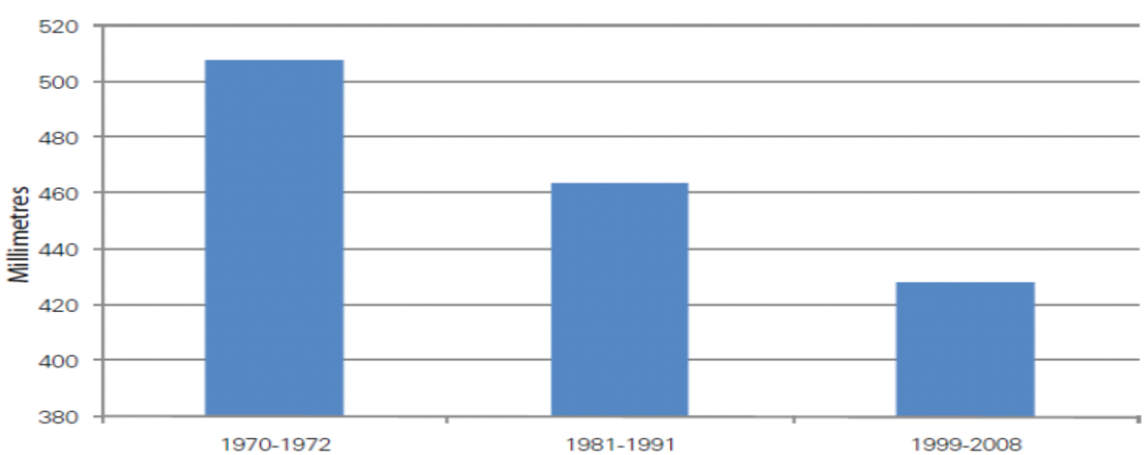


Figure 3. Average annual precipitations.

Table 2. Well location, and depth of well (Urban Core of Beijing).

Location	Construction year	Altitude (m a.m.s.l.)	Depth of well (m b.g.l.)
107	1941	47	116.50
108	1940	43	108.00
109	1940	48	106.40
111	1940	44	111.00
112	1941	50	106.60
117	1969	50	120.00
119	1942	48	66.00
122	1954	49	77.00
124	1954	47	75.00
125	1969	50	132.00
126	1954	48	95.00
127	1955	50	100.00
128	1973	49	132.50
133	1974	43	140.30
136	1973	50	100.00

Table 3. Statistic overview of hydrochemical characteristics of groundwater (2010). All units are in mg/L, except for electrical conductivity (EC, μS/cm).

Hydrochemical characteristics	Shijinshan & Hadian District			Fengtai District			Urban Core		
	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
pH	6.95	7.15	7.09	6.56	6.65	6.61	6.8	6.95	6.88
EC	650	1580	1045	1450	1790	1570	1435	1600	1540
TDS	323.78	786.11	519.81	717	891	781.33	715	801	767
Hardness	249.18	397.18	320.58	371.72	471.90	417.63	390.37	420.85	404.10
HCO_3	131.15	207.4	169.27	237.9	259.25	247.05	234.85	250.1	241.96
Cl	16.49	140.53	60.43	85.98	86.05	86.02	99.99	125.29	116.65
SO_4	65.83	139.35	91.25	123.11	127.71	126.17	92.53	167.7	130.66
NO_3	7.23	75.84	36.17	104.32	110.53	107.83	131.74	164.3	149.62
Na	17.28	69.30	38.00	67.50	106.79	81.02	59.00	98.98	83.08

areas.

FIELD AND EXPERIMENTAL INVESTIGATIONS

Sampling and analytical procedure

In May to July 2010, a groundwater sampling program was conducted at many monitoring bore in the area of study. The archival data (1960 to 2008) concerning the chemical composition of groundwater in the Region trough have been used additionally. The details of well locations, well type and depth of wells in urban core of Beijing are given in Table 2. The soil samples were also collected for mineralogical investigation. A Global Positioning System (GeoExplorer 3.0; Trimble GPS) was used for locating wells in the area of study. Physicochemical parameters, including temperature, pH, electrical conductivity (EC) and Total Dissolved Solid (TDS), were conducted in the field. Other samples were transported to the Laboratory for testing. Major components (HCO_3^-, Cl^-, $S0_4^{2-}$, NO_3^-, Ca^{2+}, Mg^{2+}, Na^+ and K^+) were measured in laboratory within 15 days of sampling. Samples were stored in the dark in a cold room at 4°C. Analysis was carried out according to established methods as follows: Alkalinity was determined by acid titration (AFNOR, 1996). Cations were analyzed with the Varian Liberty 200 Inductively Coupled Plasma - Optical Emission Spectrometer (ICP-OES) and anions were analysed with the DX300 Dionex Ion Chromatograph (EPA, 2009). The hydrochemical characteristics of groundwater in the study area were summarized as statistical overview in Table 3.

Soil phase analysis

The soil samples were collected in the study of area at a various depth of 1m to 20 m b.g.l. The soil samples were dry sieved and the < 1 mm fraction used for characterization and laboratory experiments (the > 1 mm fraction represented less than 1% of the total sample mass). Mineralogical characterization of soil samples was carried out using a D-8 advanced powder X-ray diffractometer (Bruker, Germany) and PW 1710/00 powder X-ray diffractometer

Table 4. Soil physical-chemical characteristics.

Plagioclase	Amphibole	Quartz	K-Feldspar	Calcite	Dolomite	Clay mineral amount (%)	CEC (mg/L)	TOC(%)
13.2-28.4	0.7-1.4	30.3-47.8	2.8-26.9	0.9-16.2	4.6-5.2	4.2 -23.4	3.70-7.87	4.36-10.18

(Philips, Holland). The X-ray diffractometry (XRD) patterns were obtained from the D-8 and PW1710/00 diffractometers using Cu Kα radiation equipped with a diffracted-beam monochromator in the range 5 to 60° 2θ and 10 to 60° 2θ, respectively. The voltage and current were kept at 40 kV and 30 mA, respectively. The scanning rate was kept at 2° per minute. The qualitative identification of the crystalline phases was carried out by studying the peaks in the diffractograms. The cation exchange capacity (CEC) of the soil was determined using the $BaCl_2$ Compulsive Exchange Method (Gillman and Sumpter, 1986). The total organic carbon was evaluated by titration. The results of all these analyses are shown in Table 4.

RESULTS AND DISCUSSION

Geochemical reactions modeling

The calcite content in the soils of central Beijing ranges from 0.9 to 16.2%. The pH of these soils varies from 7 to 8. The results of XRD analysis for the soil samples were as follows: the crystalline phases identified in the soil samples of Central Beijing are quartz (α-SiO_2), calcite ($CaCO_3$) as the main crystalline phase with dolomite ($CaMg(CO_3)_2$) and sodium calcium aluminium silicate anorthite - (Na, Ca) $(SiAl)_4O_8$ (Plagioclase) and K-feldspars (Table 4). Aqueous speciation modeling was carried out using PHREEQC-2 (Parkhurst and Appelo, 1999) geochemical code. The saturation indices of calcite and dolomite were computed using hydrogeochemical data of 2010. It shows that the groundwater is under saturated with calcite and dolomite, having a saturation index of -0.7856 to -0.5201 and -1.7528 to -1.2133, respectively.

The dissolution of rock by water is a very slow, thus the condition of under-saturated waters in carbonate minerals reflects a short retention time thereof in the aquifer. This is confirmed by the presence of CO_2 dissolved in the groundwater, reflecting the presence of recent groundwater aquifer. Indeed, the reaction of CO_2 hydration liberates carbonic acid attacks rocks (and Equations 2 and 3). The alteration minerals (feldspar dissolution and precipitation of silicate) lead to production of alkalinity, which consumes CO_2.

$$CO_2(gaz) + H_2O \Leftrightarrow H_2CO_3 \qquad (2)$$

$$H_2CO_3 \Leftrightarrow H^+ + HCO^-{}_3 \qquad (3)$$

In addition, the actual changes in saturation index of dolomite (SID) as a function of saturation index of calcite (SIC) help in obtaining the information on the possible relative age of groundwater (residence time), the permeability of the aquifers and the velocity of the water. To investigate this, analysis was carried out using the hydrochemical data of 2010. It is clear from the Figure 4 that, the representative points of water samples generally aligned along a regression line with linear regression equation:

$$ISD = 2.04*ISC - 0.24 \qquad (4)$$

All water wells shows an under-saturation both vis-à-vis the calcite (-0.79 < ISC < -0.36) and dolomite (-1.64 < ISD < -0.78), indicate that central part of Beijing's groundwater is relatively recent with a speed of mean circulation.

Nitrate pollution

The groundwater nitrate concentration ranges for Shijinshan and Hadian District, Fengtai District, and Urban Core and Chaoyang District were 7.23 to 75.84 mg/L, 104.32 to 110.53 mg/L, and 131.74 to 164.3 mg/L, respectively. The NO_3^- concentrations at some locations (79% of the wells) exceeded the 50 mg/L limit specified by the WHO drinking water standard. It is clear from Table 3 and figure that the region of Fengtai District and Urban core of Beijing were most affected by nitrate contamination. Figures 5 and 6 show the spatial distribution of nitrate concentration and relationship between hardness and nitrate concentrations in the area of study. It is evident that increasing nitrate concentration in groundwater hardness of groundwater increased. It can be deducted that groundwater contamination is related to the distribution of chemical water types in the study area. All water samples with high hardness were contaminated with nitrate. Nitrate concentrations of Ca subtype water are attributed to seasonal fluctuations in recharge and in plant growth. This relationship between chemical water types and contaminant concentrations is important for groundwater monitoring programs and the siting of waste-disposal facilities.

The source of nitrate in a contaminated aquifer of Beijing is critical for managing surface operations to prevent future contamination, and for evaluating exposure of human populations to nitrate. Stable isotope compositions of nitrogen and oxygen in nitrate dissolved in the groundwater could be used to determine its origin.

Once nitrates get into the groundwater, the greatest danger is for babies less than one year old. Small babies

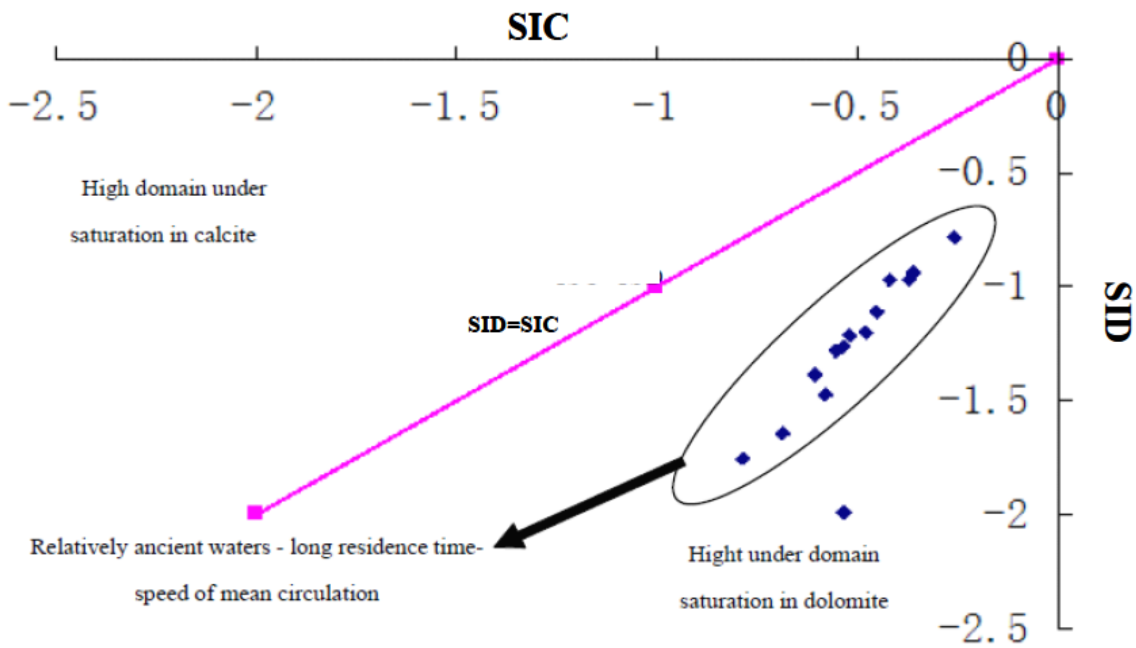


Figure 4. ISD = f (ISC) diagram of groundwater in the study area.

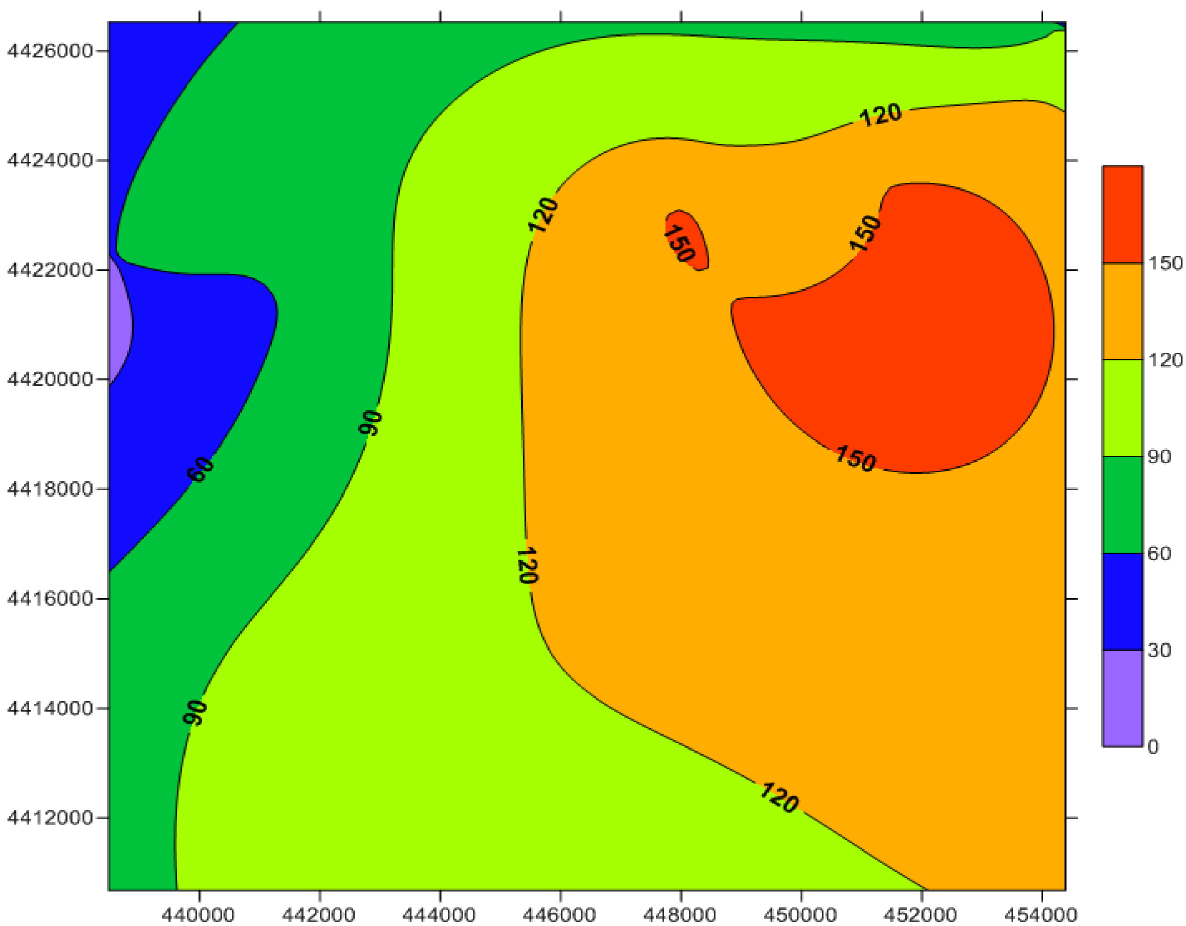


Figure 5. Spatial distribution of nitrate concentration for 2010 (mg/L).

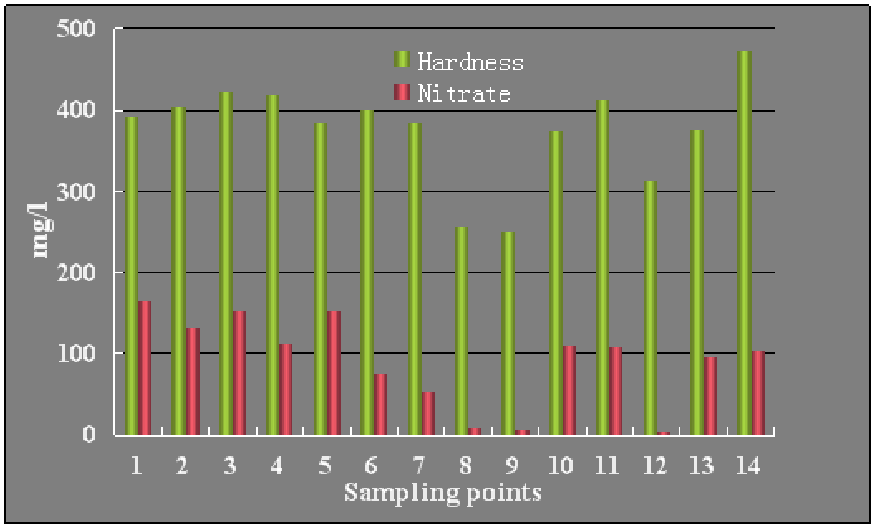


Figure 6. Relationship between hardness and nitrate concentrations.

have bacteria in their digestive tract that converts nitrate into nitrite, which is toxic. Nitrite reacts with a substance in the blood called hemoglobin. Hemoglobin is part of the red blood cell that transports oxygen to all parts of the body. When nitrites are present, hemoglobin will preferentially combine with nitrite instead of oxygen and nitrite oxidizes the hemoglobin iron from the ferrous to ferric state. This forms methemoglobin, which is unable to bind oxygen to carry to tissues. As the amount of methemoglobin increases, the amount of oxygen in the blood decreases, eventually causing internal suffocation, mild cyanosis, reduced level of consciousness, and death (Walton, 1951; WHO, 1996; U.S. EPA, 1997).

The most common symptom of nitrate poisoning in babies is a bluish color to the skin, particularly around the baby's eyes and mouth. The blood will also turn a chocolate-brown color, which reflects the lack of oxygen. These symptoms of nitrate toxicity are commonly referred to as the "blue-baby" syndrome.

Groundwater exploitation and evolution of hardness

The estimated mean annual recharge is 3.8 Mm^3 and the exploitable resource is 2500 Mm^3. Due to a sharp increase of the demand for domestic, industrial and agricultural water since 1970, the yield exceeded the permissive annual maximum of 2500 Mm^3 upsetting the dynamic equilibrium of groundwater. Local cones of depression in the centralized exploitation areas have been enlarged year by year and have become connected, forming a regional depression cone whose area attained 1000 km^2 in 1980. The groundwater table declines at an annual rate of 0.5 to 1.5 m (maximum, 2 m). In the center of the cone the water table is now more than 40 m below the ground surface, with the result that the first confined aquifer and phreatic layers have dried up. The third confined aquifer which is more than 150 m deep is also developed, and its level declines at an annual rate of 1.5 to 2.0 m per year. Because of the decline of the regional water table over a large area the hydrodynamic conditions have changed. For example, in the section from Jianguomen to Bawangfen, the hydraulic gradient of groundwater has increased from 1.70 to 1.90% before 1974 to 3.70 to 4.34% inducing the lateral entry of hard groundwater into the exploited areas and increasing the hardness of Beijing groundwater each year.

Besides the pollution directly caused by domestic sewage and industrial waste, it may be shown that in many cases the pollution is due to intense exploitation of groundwater. In Beijing, the total hardness of groundwater was 230 to 500 mg/L in 1960s, but now it exceeds 650 mg/L and even 800 mg/l in some places, whereas the state standard for drinking water is only 450 mg/L. The area where the total hardness of the groundwater has risen beyond the national standard of 450 mg/L for potable water had reached 217 km^2 by 1980 (1.6 times greater than in 1970). In consequence, some production wells have had to be abandoned. Preliminary studies have shown that the increase of hardness is related to the large scale exploitation of groundwater. That is, the lowering of the water table in the exploited aquifer induced more highly mineralized underground water to penetrate into the exploited aquifer, with a resultant increase in hardness. Therefore, limitation of the use of groundwater is extremely important to prevent its over-extraction and deterioration. The increased hardness not only does harm to public health, but also influences the normal production from pharmaceutical factories, wineries and other enterprises.

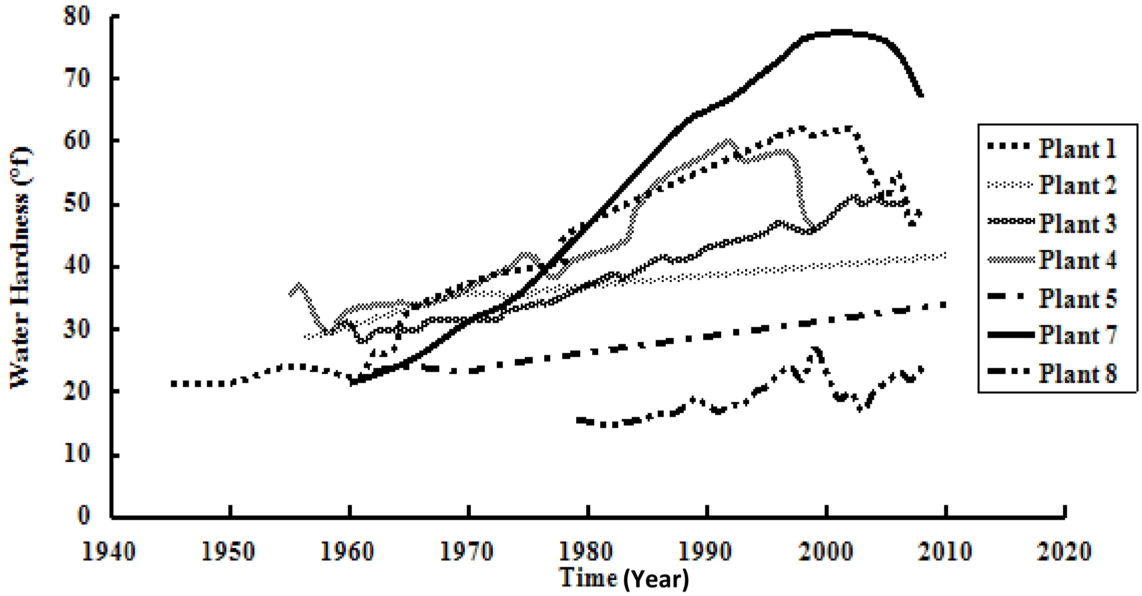


Figure 7. Changes of hardness with time.

In addition, the salinity of the water in the shallow layers is very high. As a result, during the course of the vertical and lateral groundwater recharge by precipitation in the exploited areas, exchanges occur between Na and Ca, Mg resulting in Ca, Mg entering the water, further increasing the hardness of groundwater in the exploited areas. Thus, the chemical components of shallow groundwater in the urban district changed from Cl, HCO_3-Ca Mg, Na into Cl, HCO_3 Ca Mg.

The rates of increase of the hardness of groundwater in the zone of intensive withdrawal were rapid at first in the east then fell off, whilst in the west the reverse has been observed, slow at first followed by a sharp increase. Two factors are responsible for this situation. In the early of 1950's, water from the polluted water table flowed down into confined aquifers 2 and 3, increasing the hardness of the water rapidly from 230 to 450 mg/L, well over the accepted limit for drinking water. After 1965, the rate of increase of hardness in confined aquifers 2 and 3 decreased gradually because of depletion of the water table aquifer and concurrent fluctuations of water levels in the water table aquifer and confined aquifers 2 and 3. The hardness at Water Plant 5, near the edge of the regional cone of depression, hardly increased at all because of the presence of a thick and stable clay bed between the aquifers and because the amount of lateral recharge is considerable. In the western water table aquifer region, the hardness increased slowly because withdrawals were not excessive between 1950 and 1960. But in the 1970's the water table started to fall as result of overdrafts. The rapid hydraulic cycle, the increase of thickness of the non-saturated zone and the deposition of soluble salts moving down from the polluted ground, are the principal causes for the increase of the total dissolution and hardness of the water table in general and particularly of wells at Water Plant 7, located in the area of thin overburden polluted by irrigation water (Figure 7).

High degrees of hardness in groundwater affected urban areas first and suburban areas later, in the east first and the west afterwards and the shallow before the deep aquifers. Groundwater hardness over 720 mg/L was concentrated in the small southern urban area of Tiantan-Longtanhu which has a long history of pollution. With the development of groundwater in the eastern suburb, high groundwater hardness has extended northeastward, reaching a peak in the mid 1990's. By the 2000's with the control of the city environment and the depletion of the water table aquifer, the growth of hardness was brought under control and there has even been a small decrease of its value as well as of the area affected (Figure 8).

Before 1946, the hydrogeochemistry data are scarce, so it is very difficult to know exactly when hardness began rising in groundwater. Due to over-withdrawal by the Water Plants, the area of hard water now extends over the entire city area (Figure 8). By 1980, the area affected by hard water was 15 times greater than in 1946. Except in the case of Water Plants 5 and 8 which have good lateral recharge, the degree of hardness often exceeds the accepted standard for drinking water, increasing the cost of water treatment and putting some wells out of production. In Figure 8, based on the Hardness Classification of Ragunath (1987) the period of 1960 shows that 50% of samples fall under very hard category and 50% of samples under hard category. From 1970 all samples fall under very hard category.

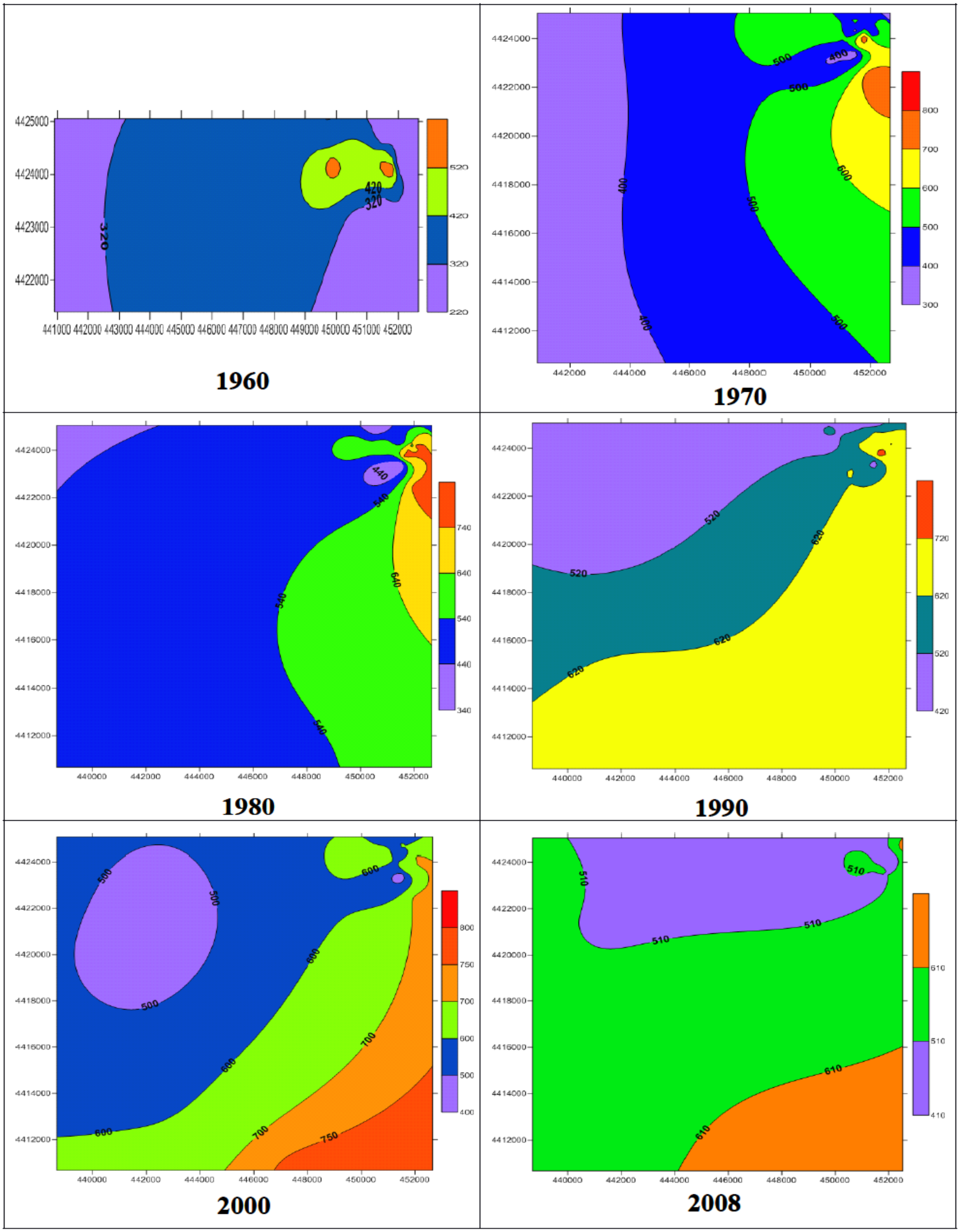


Figure 8. Total hardness in the area of study.

Conclusion

Nitrate contamination of the world's groundwater supply poses a serious human health threat. High nitrate levels found in drinking water have been proven to be the cause for numerous health conditions across the world. If we intend to provide for the future survival of man and life on planet earth, we must take action now to assure the quality of one of our most precious resources, our groundwater supply. As a result of environmental studies, water quality monitoring and other investigations, the calcite and dolomite contents of strata around Beijing are known to be generally 6 to 15%, with the highest values about 21%. Following environmental pollution, reactions occurred between sewage and the surrounding medium as the sewage infiltrated the ground. As a consequence of a series of physico-chemical and biochemical reactions, including dissolution, ion-exchange and reactions involving organic compounds, the Ca and Mg in the infiltrating solutions has increased. The solutions have infiltrated into aquifers and increased the hardness of the groundwater by 9 mg/l as $CaCO_3$ per year; overpumping of groundwater has accelerated the process. If measures were adopted to prevent or control pollution, the quality of groundwater in the Beijing area would recover and gradually improve.

ACKNOWLEDGEMENT

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