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Characteristics of Heavy Metal Contents and Health Risk Assessment Concerning Rural Groundwater in Suzhou City, China

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Abstract: 70 groundwater samples were collected from the rural area in Suzhou, Anhui province, and seven heavy metals (including Fe, Mn, Cr, Cd, Cu, Pb and Ni) in each sample were tested. Based on the analysis of heavy metal content characteristics, we carried out quality evaluation on the groundwater samples with Nemerow composite index method, and conducted health risk assessment (HRA) with the recommended model by the United States Environmental Protection Agency (USEPA). The results show that: (1) the descending order of the average concentration of heavy metals in the samples is Mn > Fe > Ni > Pb > Cu > Cr > Cd. Mn and Ni concentrations in excess of the concentration requirements set by the Standards for Drinking Water Quality (GB5749-2006) are 32.85% and 5.71% that of and 2.97 times and 2.28 times higher than the set values, respectively. (2) The comprehensive evaluation score of the groundwater samples according to Nemerow composite index method is 0.2818～2.1292, with the mean value of 0.6800, and the ground water quality level is "favorable". (3) For chemical non-carcinogens (Fe, Mn, Cu, Pb and Ni) that are ingested through the mouth, the descending order of their health risk levels is Pb>Cu>Mn>Ni>Fe, and all of their risk scores are below the recommended maximum acceptable value ($5.0\times10^{-5}a^{-1}$) of the International Radiation Protection Association(IRPA) and below the recommended health risk standard (1×10^{-4} a^{-1}) of USEPA. This means that the ingestion of the tested groundwater will basically not pose significant hazards to exposed populations. The mean health risk scores of carcinogens (Cd and Cr) by mouth is $9.96\times10^{-7}a^{-1}$ and $1.07\times10^{-5}a^{-1}$, respectively. The risk score of Cr is 10.7 times larger than the recommended maximum acceptable value ($1\times10^{-6}a^{-1}$) by the Swedish Environmental Protection Agency, the Ministry of Housing, Spatial Planning and the Environment (Netherlands), and the British Royal Society. Therefore, the plethora of Cr renders it the representative pollutant of the research area such that priority should be given to it.

***Keywords:** Suzhou, groundwater, heavy Metals, water Quality analysis, health risk*

1. Introduction

Compared to surface water, ground water is characterized by wide distribution, high water quality, steady change, easy access, and unlikelihood of being contaminated as well, and has been extensively used in daily life and producing activities [1]. According to statistics, groundwater supply occupies one third of the total water supply in China. Among the 655 cities in China, more than 400 ones use groundwater as the source of drinking water [2-3]. Recently, groundwater quality has continued deteriorating as a result of undue groundwater exploitation, sewage permeation, and various industrial and agricultural activities [4-5]. Data shows that nationally, 25% groundwater has been polluted, 35% groundwater source is below standard, and over 300 cities are in short water supply caused by groundwater contamination. About 54% groundwater of plain areas fails to comply with the Standards for Drinking Water Quality, and more than half of urban groundwater is seriously polluted [6-7]. As environmental pollutants and potential toxic pollutants, highly-stable, accumulative, poisonous heavy metals greatly endanger human health [8-9]. Heavy metals enter nature water through different accesses, such as sewage emission, coal mining, agricultural non-point source pollution, and atmospheric precipitation. Aquatic animals and plants carry the heavy metals by ingestion, and endanger human health directly or indirectly when they enter human body through the mouth or food chains [10-12]. In terms of the research area in the paper, there has been a small amount of water quality evaluation and HRA on sources of urban water and groundwater of mining areas [13-14], but little on rural groundwater. Given that rural groundwater quality impacts on daily life greatly, the development of rural groundwater quality analysis and HRA is of great significance to guaranteeing water supply safety and human health.

Suzhou city, in the north of Anhui province, is located at 116°09'-118°10'E, 33°18'-34°38'N. It is an area where four Chinese provinces (Anhui, Jiangsu, Shandong, and Henan) intersect. Governing four counties (Dangshan, Xiaoxian, Lingbi, Sixian) and one district (Yongqiao district), Suzhou city has an area of 9,787 square km, and a population of

6,516,600. The city is geologically situated at the warm temperate zone, and has a semi-humid monsoon climate with the average annual temperature of 14~14.5°C. The annual precipitation amount in the city is in the range of 774mm to 895mm, being produced mainly in May-September. Almost all of the water resources for daily, industrial, and agricultural use come from ground water [17]. The total number of water resources is 3.48 billion m3, and the water resources per capita is 602m3, which means that Suzhou city is a city of severe water shortage [18-19]. With abundant coal resources, the city is one of the 13 national planned large coal bases [20]. The proved reserves of its coalbed methane and coals are 60 billion cubic meters and 6 billion ton, respectively.

2. Materials and methods

2.1 Sample collection and analysis

The paper collected 70 groundwater samples (1L per one) from the well of local villagers during Sept. 2013 and Oct. 2013. Each of the sample was put into a polyethylene bottle that had been rinsed by deionized water, and was taken back to the laboratory within 24 hours. During the laboratory test, the samples first underwent microfiltration with a particle size of 0.45μm. Then, purified HNO_3 was added to the sample until the pH decreased to or below 2. Next, TAS-990 atomic absorption spectrophotometer was used to analyze seven heavy metals (Fe, Mn, Cr, Cd, Cu, Pb and Ni). Qualitatively, the flame test was done on Fe and Mn, and the rest metals were tested by GFASS; while external standard method was used for all the metals for quantitative results. It turned out that the relative coefficients for all the 7 calibration curves were higher than 0.998.

2.2 Evaluation approaches

There are numerous aquatic environment evaluation approaches at home and abroad, such as Nemerow composite index method, fuzzy mathematic evaluation method, gray correlation method, and artificial neural network [21-22]. Among them, the Nemerow composite index method that is recommended by the Standards for Ground Water Quality (GB/T 14848-93) was used as a basis for groundwater quality evaluation in the paper, and the recommended HRA method by USEPA was used hereby.

2.2.1 Nemerow composite index method

The Nemerow composite index method reflects contamination laws of various pollutants, and gives consideration to the water quality parameter with the greatest pollution impact, thus well displaying the excessive amount of constituents [24]. By taking comprehensive water usage into account, this method provides certain practical value. The steps of this method are: first, evaluate the quality levels of each single constituent according to the Standards for Ground Water Quality (GB/T14848-2007)-ClassⅢ by constituent (Table 1), and determine the evaluation scores Fi of the constituents; second, calculate the comprehensive evaluation score F, by which the groundwater quality level is obtained (Table 2) [26]. Equation (1) is the formula of F:

$$F=\sqrt{\frac{F_{\max}^2+\overline{F}^2}{2}}=\sqrt{\frac{\left(\frac{Ci}{Coi}\right)_{\max}+\frac{1}{n}\sum_{i=1}^{n}\left(\frac{Ci}{Coi}\right)^2}{2}} \quad (1)$$

Where: F-the comprehensive pollution index; n-the number of pollution factors under evaluation; i-a single factor; C_i-the practical measured value of the water quality factors; C_{0i}-the standard Class III value of the *i*th factor; $\overline{F}$ -the average value of F_i; F_{max}-the maximum value among F_i.

***Table 1:** 《Standards for Ground Water Quality》 (GB/T14848-2007) - Class Ⅲ by constituent (mg/L)*

Fe	Mn	Cr	Cd	Cu	Pb	Ni
≤0.3000	≤0.1000	≤0.0500	≤0.0100	≤1.0000	≤0.0500	≤0.0500

***Table 2:** Ground water quality level classifications*

Level	excellent	favorable	good	Relatively poor	poor
F	＜ 0.80	0.80～2.50	2.50～4.25	4.25～7.20	＞ 7.20

2.2.2 HRA

By connecting pollutants with human health, HRA quantitatively describes risks of pollutants on human health [27-28]. Compared to traditional water quality level evaluation systems, HRA reflects the potential risks of all kinds of aquatic pollutants on human health more visually [29]. The paper employed the recommended HRA method by the USEPA, and established a corresponding model to evaluate the health risks of heavy metals in groundwater samples.

The health risks of the chemical carcinogens (represented by i) and the chemical non-carcinogens (represented by j), which arise upon human kinds drinking the groundwater, are calculated according to Equation (2) and Equation (3) as follows [30-32].

$$R_i^c=\frac{1-\exp(-D_i q_i)}{L} \quad (2)$$

$$R_j^n=\frac{D_j\times10^{-6}}{RfD_j\times L} \quad (3)$$

Where R_i^c and R_j^n denote the annual average carcinogenic risks of i and j by mouth, respectively,a^{-1}; D_i and D_j represent the daily exposed dosage of i and j by mouth per unit of human body weight, mg/(kg·d); q_i is the carcinogenic strength coefficient of i, mg/(kg·d); RfD_j is the reference dosage of j, mg/(

kg·d) ;and L is the average life of the exposed population in the research area [33], a.

D_i and D_j are calculated according to Equation (4).

$$D_{i/j} = \frac{w \times C_{i/j}}{A} \quad (4)$$

Where w denotes the daily water intake amount, whose general value of adult humans is 2.2 L/d; $C_{i/j}$ is the mass concentration of i by mouth, mg/L; and A is the body weight per capita, which is taken as 70kg [34] in adults.

Restricted by levels of current research, the paper presumed that there is no mutual antagonism or synergistic relations between the toxic effects of the different heavy metals on human health [35]. The total health risk, which is obtained by adding the total health risk of i by mouth (R^c) to the total health risk of j by mouth (R^n) is computed according to Equation (5) and Equation (6).

$$R_{Total\ \ drinking\ \ water} = R^c + R^n \quad (5)$$

$$R^c = \sum_{i=1}^{m} R_i^{\ c} \qquad R^n = \sum_{j=1}^{k} R_j^{\ n} \quad (6)$$

In line with relative data from International Agency for Research on Cancer (IARC) and WHO, Cd and Cr belong to chemical carcinogens, while Mn, Cu, Zn, Pb and Ni belong to chemical non-carcinogens (Table 3).

Table 3: *Values of q_i and RfD_j of model parameters via drinking water*

chemical carcinogens	q_i / (mg/(kg•d))	chemical non-carcinogens	RfD_j /(mg/(kg•d))
Cd	6.1	Mn	1.4×10^{-1}
Cr	41	Cu	5.0×10^{-3}
		Fe	3.0×10^{-1}
		Pb	1.4×10^{-3}
		Ni	2.0×10^{-2}

3. Results and discussions

3.1 The characteristics of heavy metal contents

The paper compared the tested contents of the heavy metals with the reference contents in the Standards for Drinking Water Quality (GB5749-2006), aiming to provide a better reference for rural residents in Suzhou city. The results are shown in Table 4.

Table 4: *The characteristics of heavy metal contents in rural groundwater of Suzhou city*

Item	Unit	Fe	Mn	Cr	Cd	Cu	Pb	Ni
the Standards for Drinking Water Quality	mg/L	0.3000	0.1000	0.0500	0.0050	1.0000	0.0100	0.0200
sampling point	/	0	23	0	0	0	1	4
Maximum value	mg/L	0.0330	0.2970	0.0035	0.0024	0.1008	0.0103	0.0455
Minimum value	mg/L	0.0005	0.0390	0	0	0.0003	0.0020	0
Mean value	mg/L	0.0143	0.0932	0.0006	0.0004	0.0039	0.0044	0.0072
Maximum excess times	/	0.1100	2.9700	0.0700	0.48	0.1008	1.0300	2.2750
Minimum excess times	/	0.0017	0.3900	0	0	0.0003	0.2000	0
Excess rate	%	0	32.85	0	0	0	1.43	5.71

For the groundwater samples, the contents of Fe, Mn, Cr, Cd, Cu, Pb and Ni are in the range of 0.0005～0.0330mg/L,0.0390～0.2970mg/L,0～0.0035 mg/L,0～0.0024 mg/L,0.0003～0.1008 mg/L,0.0020～0.0103 mg/L, and 0～0.0455 mg/L, respectively. The descending order of the average concentrations for each of the seven heavy metals is Mn>Fe>Ni>Pb>Cu>Cr>Cd. The result of Mn>Fe in the paper and the result of Fe>Mn in the research on heavy metal contents of shallow groundwater in Huainan city by He Xiaowen et al. [36] are contradictory. The result of the studies on groundwater quality in Suzhou city by Li Lei et al. [37] showed that the content of Fe was high, which agreed with the result in the paper. High-content Fe in groundwater poses enormous hazards to daily life, industrious production, and agricultural production, therefore relative governmental departments are supposed to pay attention to the issue.

In addition, compared to the Standards for Drinking Water Quality (GB5749-2006), the tested excess rate of Mn (32.85%) and Ni (5.71%) is separately 2.97 times and 2.28 times that of the required values. Mn has the largest excess point of 23 and the highest excess rate, which agrees with the result of research on groundwater in Zhongshan city, Guangdong province by Liu Junke et al.[38].

3.2 Comprehensive water quality evaluation

According to the formula in Nemerow composite evaluation method, the results of comprehensive evaluation on the groundwater quality were obtained, as shown in Table 5. Table 6 is the statistics of Suzhou groundwater quality levels and its percentages.

As can be seen from Table 5 and Table 6, F is 0.2818～2.1292, with the mean value of 0.6800. 53 out of 70 sampling points have excellent water quality, and the rest sampling points have favorable water quality, which means that the main water quality of the search areas is excellent. This result agrees with that of the groundwater during dry seasons and wet seasons in Chenzhou city, Hunan province which was tested by Xu Bingbing et al. [39], and surpasses the result that F is 3.63 and that the main water quality is "favorable" in the research on groundwater quality in Wuxi city, Jiangsu province which was conducted by Gu Zhonghua et al. [26].

Table 5: *The results of comprehensive evaluation on the groundwater quality, Suzhou city (mg/L)*

Sampling point	Pollution value of a single index							Nemerow composite index evaluation value
	Fe	Mn	Cr	Cd	Cu	Pb	Ni	F
Maximum value	0.1101	2.9700	0.0704	0.2354	0.1008	0.2068	0.9093	2.1292
Minimum value	0.0016	0.3900	0.0004	0.0009	0.0003	0.0404	0.0007	0.2818
Mean value	0.0478	0.9317	0.0125	0.0390	0.0039	0.0875	0.1438	0.6800

Table 6: *Suzhou groundwater quality levels and its percentages*

Level	excellent	favorable	good	Relatively poor	Poor
F	＜ 0.80	0.80～2.50	2.50～4.25	4.25～7.20	＞ 7.20
sampling point	53	17	0	0	0
percentage	75.7%	24.3%	0.0%	0.0%	0.0%

3.3 Results of the health risk assessment

Based on the mathematic model of health risk evaluation and relative parameters, the paper computed the annual health risk per capita and the total health risk caused by the chemical carcinogens and the chemical non-carcinogens by mouth, as shown in Table 7.

Table 7: *The annual health risk per capita and the total health risk caused by the chemical carcinogens and the chemical non-carcinogens by mouth (a^{-1})*

	chemical non-carcinogen					chemical carcinogen		Total health risk
	Fe	Mn	Cu	Pb	Ni	Cr	Cd	
The maximum risk	4.61×10^{-11}	8.89×10^{-10}	8.45×10^{-9}	3.09×10^{-9}	9.53×10^{-10}	6.03×10^{-5}	6.02×10^{-6}	6.64×10^{-5}
The minimum risk	6.50×10^{-13}	1.17×10^{-10}	2.43×10^{-11}	6.05×10^{-10}	7.12×10^{-13}	3.78×10^{-7}	2.40×10^{-8}	4.02×10^{-7}
The mean risk	2.00×10^{-11}	2.79×10^{-10}	3.27×10^{-10}	1.31×10^{-9}	1.51×10^{-10}	1.07×10^{-5}	9.96×10^{-7}	1.17×10^{-5}

As can be seen from Table 7, the average values of health risks of Fe, Mn, Cu, Pb and Ni by mouth are 2.00×10^{-11} ,2.79×10^{-10}, 3.27×10^{-10}, 1.31×10^{-9} and 1.51×10^{-10},respectively, Pb>Cu>Mn>Ni>Fe. According to the research result of groundwater in a certain city by Li Shanshan et al. [28], the health risk magnitude of chemical non-carcinogens is 10^{-11}-10^{-9},and the main pollutant is Pb, which agrees with the research result in the paper. The risk score of chemical non-carcinogens in the research area is far below the recommended maximum acceptable value ($5.0\times10^{-5}a^{-1}$) of the IRPA and below the recommended health risk standard (1×10^{-4} a^{-1}) of the USEPA. This means that the ingestion of the tested groundwater will basically not pose significant hazards to exposed populations.

The mean health risk scores of carcinogens (Cd and Cr) by mouth is $9.96\times10^{-7}a^{-1}$ and $1.07\times10^{-5}a^{-1}$, respectively. The risk score of Cr is 10.7 times larger than the recommended maximum acceptable value ($1\times10^{-6}a^{-1}$) by the Swedish Environmental Protection Agency, the Ministry of Housing, Spatial Planning and the Environment (Netherlands), and the British Royal Society. This result agrees with the result that the health risk level of Cr exceeded that of Cd in the research on groundwater contamination in areas of Yangtze Delta River by Huang Lei et al. [40]. According to a study on heavy metals of drinking water in Baoding city, Hebei province by Yang Yang et al. [41], Cd posed the maximum carcinogenic risk to the exposed population in the upgraded downtown of Baoding city, which is different from the research result in the paper; whereas the contents of Cd in both the research areas were lower than the recommended maximum acceptable value ($5.0\times10^{-5}a^{-1}$) of the IRPA. In addition, the health risk magnitude of chemical carcinogens is 2-7 larger than that of chemical non-carcinogens, which means that chemical carcinogens are the prior targets for prevention and control, and that relative governmental departments should emphasize on them.

Practically, the obtained health risk of heavy metals is an underestimate, because the paper merely conducted an ingestion-based health risk assessment on seven heavy metals, and excluded other toxic constituents and exposed approaches (skin exposure, and inhalation, for example) in the research. What's more, the health risk by mouth is closely linked to lifestyles, living habits, and career types. However, the paper computed the total health risk by purely adding the total health risk of chemical carcinogens by mouth to the total health risk of chemical non-carcinogens by mouth, without giving consideration to the possible mutual antagonism or synergistic relations between the toxic effects of the different heavy metals on human health. The uncertainty of health risk assessment rendered the reference dosage and the carcinogenic strength coefficient uncertain. All in all, there are many research points that remain to be further studied.

4. Conclusions

1) The descending order of the average concentration of heavy metals in the samples is Mn > Fe > Ni > Pb > Cu > Cr > Cd. Mn and Ni concentrations in excess of the concentration requirements set by the Standards for Drinking Water Quality (GB5749-2006) are 32.85% and 5.71% that of and 2.97 times and 2.28 times higher than the set values, respectively.

2) The comprehensive evaluation score of the groundwater samples according to Nemerow composite index method is 0.2818～2.1292, with the mean value of 0.6800, and the ground water quality level is "favorable". This means that the general groundwater quality in rural areas of Suzhou city is superior.

3)According to the results of the health risk assessment, for chemical non-carcinogens (Fe, Mn, Cu, Pb and Ni) that are ingested through the mouth, the descending order of their health risk levels is Pb>Cu>Mn>Ni>Fe. The mean health risk scores of carcinogens (Cd and Cr) by mouth is $9.96\times10^{-7}a^{-1}$ and $1.07\times10^{-5}a^{-1}$, respectively. The risk score of Cr is 10.7 times larger than the recommended maximum acceptable value ($1\times10^{-6}a^{-1}$) by the Swedish Environmental Protection Agency, the Ministry of Housing, Spatial Planning and the Environment (Netherlands), and the British Royal Society. Therefore, the plethora of Cr renders it the representative pollutant of the research area such that priority should be given to it.

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