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Research Article

Human Health Risk Assessment of Heavy Metals in Groundwater in the Luan River Catchment within the North China Plain

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The Luan River catchment within the North China plain has been famous for the development of its iron mineral resources since the 1950s. At the same time, it is also the main grain-producing area, known as the granary of eastern Hebei Province. Groundwater plays an important role in this region, and thus, it is imperative for us to improve our understanding of the heavy metal groundwater contamination in this catchment. Therefore, a total of 144 groundwater samples were collected for chemical analysis from 16 operational private wells of local residents in the study area, over eight consecutive periods from December 2016 to May 2017. Each shallow groundwater sample was analyzed for 39 heavy metals including among others, As, B, Ba, Be, Cd, Cr, Cu, Fe, Li, Mn, Mo, Ni, Sb, Se, Sn, Sr, V, and Zn. However, subsequent analyses only focused on three heavy metals (Cd, Cr, and Ni) that exceeded the Groundwater Quality Standard III. Spatial and temporal variations of Cd, Cr, and Ni in the shallow groundwater were analyzed. Cr was found to have the highest concentrations, followed by Ni and Cd. A human health risk assessment was conducted where carcinogenic risks and Hazard Quotients (HQs) were evaluated separately. The results indicate that both the carcinogenic risks and HQs of Ni and Cr are higher than the recommended standard value. Therefore, the prevention and control of heavy metal contamination in the Luan River catchment should focus on Ni and Cr.

1. Introduction

Less than 3% of the Earth's water resources are freshwater, and only one-hundredth of a percent of this is adapted for human consumption [1]. Groundwater makes up 20% of the world's fresh water supply [2, 3]. There is no doubt that groundwater plays a significant role in strengthening the economic growth of developing countries [2, 4] and where it is also indispensable for drinking, domestic use, industry, and agriculture [5] and therefore directly or indirectly influences daily life. Groundwater is also an essential component of the ecological and geological environment and greatly impacts biological growth and human life [6]. However, industrial development and economic growth in developing countries have also resulted in the heavy metal contamination of groundwater, which has become a serious global problem [7–10]. As a result, research studies have been focusing on quantifying heavy metals and their effects in aqueous environments [8, 11–14]. Groundwater in the Luan River catchment within the North China plain has also been polluted by industrial, agricultural, and domestic wastewater. Luan River water quality has deteriorated and become a serious threat to the local ecological environment, including food crops, vegetables, soil, the vadose zone, and aquifer layers. In turn, the deteriorated local ecological environment can result in considerable health risks to humans and animals. Consequently, how to effectively control and remediate ecological environments affected by heavy metal pollution has become a key issue that directly affects the healthy and sustainable development of the economy.

Among the pollutants that can affect water resources, heavy metals are paid more attention due to their high toxicity at low concentrations [15]. In case of uncontrolled heavy metals in the environment, they can lead to health effects such as poor growth and development, cancer, nervous system damage, and even death [15]. Heavy metals in drinking water are known to be toxic and a serious threat to human health, based on common and widespread documented evidence from many parts of the world. The toxicities of select heavy metals are given in Table 1. Although some heavy metals (e.g., Cu, Mn, and Cr) are essential for humans, their presence in excess amounts may be toxic. In addition, other metals (e.g., As, Hg, Cd, and Pb) are highly toxic at very low concentrations with no known benefits for human health [2, 16–18]. The heavy metals mentioned above cannot be eliminated from aqueous systems and are often recycled via physiochemical and biological processes and continue to pose adverse risks to human health and aqueous ecosystems [19]. Characterizing the heavy metal content of groundwater is necessary in order to understand the sources, fate, and potential health risks of heavy metals [20]. It is thus imperative for us to improve our understanding of heavy metal groundwater contamination in the Luan River catchment within the North China plain. Therefore, the objectives of this study were to (1) investigate the distribution patterns of heavy metals in groundwater, (2) determine the possible sources of heavy metals in groundwater, and (3) assess the potential risks to public health. A risk assessment model was used to calculate Hazard Quotients (HQs) and carcinogenic risks to assess potential risks to human health associated with select metals. The research outcome of the present work is expected to provide necessary information for implementing the necessary precautions and remedial measures, for sustainable development, and effective groundwater management in the Luan River catchment in North China plain.

2. Study Area and Sampling

2.1. Study Area. The Luan River basin is mainly located within the eastern portion of Hebei Province between a latitude of 39°44′–42°44′N and a longitude of 115°33′–119°36′ E. It provides a considerable amount of water for agricultural, domestic, industrial, and other purposes. This region has been famous for the development of its iron mineral resources since the 1950s. In addition, it is also a main grain-producing area, known as the granary of eastern Hebei Province. The water resources are essential for mining the iron mineral resources and for agricultural activities associated with the granary. With the rapid industrialization, agriculturalization, and urbanization of the eastern portion of Hebei Province, a series of water problems have emerged, including severe water pollution and water shortages. The study area is located in the lower part of the Luan River catchment in the southeastern portion of Hebei Province and the northwestern region of the Bohai Sea and spreads between 39°30′–39°40′N in latitude and 118°50′–119°00′E in longitude and encompasses an area of approximately 150 km^2 (Figure 1).

The area generally slopes from northwest to southeast as indicated by the direction of rivers flowing through it. The slope gradient in the study area is 0.1%–0.2%, and thus, the topography is relatively flat. To our best knowledge, the study area has a typical warm temperate semihumid continental monsoon climate, affected by maritime polar air masses or denaturing tropical marine air masses, with high temperatures and abundant rainfall in the summer and low temperatures and less rainfall in the spring. Under the influence of north wind from high-latitude inland areas, the climate is cold and dry in the winter. The air temperature in this region ranges from -11°C to 30°C, with an average annual temperature of 10.1–11.0°C, and the total annual rainfall ranges from 747 to 772 mm [21, 22]. The proportion of yearly evapotranspiration from April to June is about 47.7% [23]. According to the lithology and sedimentary sequences of the study area, this region can be divided into unconfined and confined aquifers. The unconfined aquifer has a close hydraulic connection with surface water, and therefore, the Luan River and precipitation are the main sources of recharge for this aquifer. The aquifer depth ranges from 60 to 70 m, and the water table decreases from 2.7 m near the Luan River to between 7.6 and 13 m in the southwestern and southeastern parts of the study area. This area has an aquifer group that includes three layers. The burial depth of the first aquifer, which consists of silt sand and fine sand, is 6–10 m, with a thickness of 3 m. The burial depth of the second aquifer is 25–30 m, with a thickness of 10 m. The third aquifer is a semiconfined aquifer occurring at depths below 60 m with a thickness of 10–20 m [7, 24].

2.2. Samples. Groundwater samples were collected to investigate the distribution and possible sources of heavy metals in groundwater in the Luan River catchment within the North China plain and to calculate HQs and carcinogenic risks to assess the potential harm to public health. A total of 144 groundwater samples were collected from the study area from 16 operational private wells of local residents, over eight consecutive periods from December 2016 to May 2017. The geographical location of the sampling sites is shown in Figure 1. The sampling sites are distributed along both sides of the Luan River. The number of sampling sites north and south of the Luan River is different, with the area to the south having two additional sites. Prior to sampling, the bottles were rinsed several times with deionized water and then three times with the groundwater being sampled in order to minimize the potential for contamination. Water samples from bore wells were collected after pumping out water for about 10 min to remove stagnant water. Each sample was collected in a 2.5 L clean and sterile polyethylene drum that was numbered and labeled with a different site code. Groundwater samples were transported as soon as possible to the Institute of Hydrogeology and Environmental Geology for heavy metal determination. A total of 39 heavy metals were analyzed including among others, As, B, Ba, Be, Cd, Cr, Cu, Fe, Li, Mn, Mo, Ni, Sb, Se, Sn, Sr, V, and Zn. The water samples were filtered and acidified at the laboratory and then analyzed by inductively coupled plasma mass spectrometry (ICP-MS, NexION300D) for heavy metal concentrations. Safety measures were applied at every stage of sample handling, starting from sample collection to storage, transportation, and final analysis in order to minimize contamination and ensure the precision and accuracy of the measurements.

Table 1: The toxicities of some heavy metals [15].

Heavy metals	Toxicities
Arsenic	Skin manifestations, visceral cancers, and vascular disease
Cadmium	Kidney damage, renal disorder, and human carcinogen
Chromium	Headache, diarrhea, nausea, vomiting, and carcinogenic
Copper	Liver damage, Wilson disease, and insomnia
Nickel	Dermatitis, nausea, chronic asthma, coughing, and human carcinogen
Zinc	Depression, lethargy, neurological signs, and increased thirst
Lead	Damage the fetal brain; diseases of the kidneys, circulatory system, and nervous system
Mercury	Rheumatoid arthritis and diseases of the kidneys, circulatory system, and nervous system

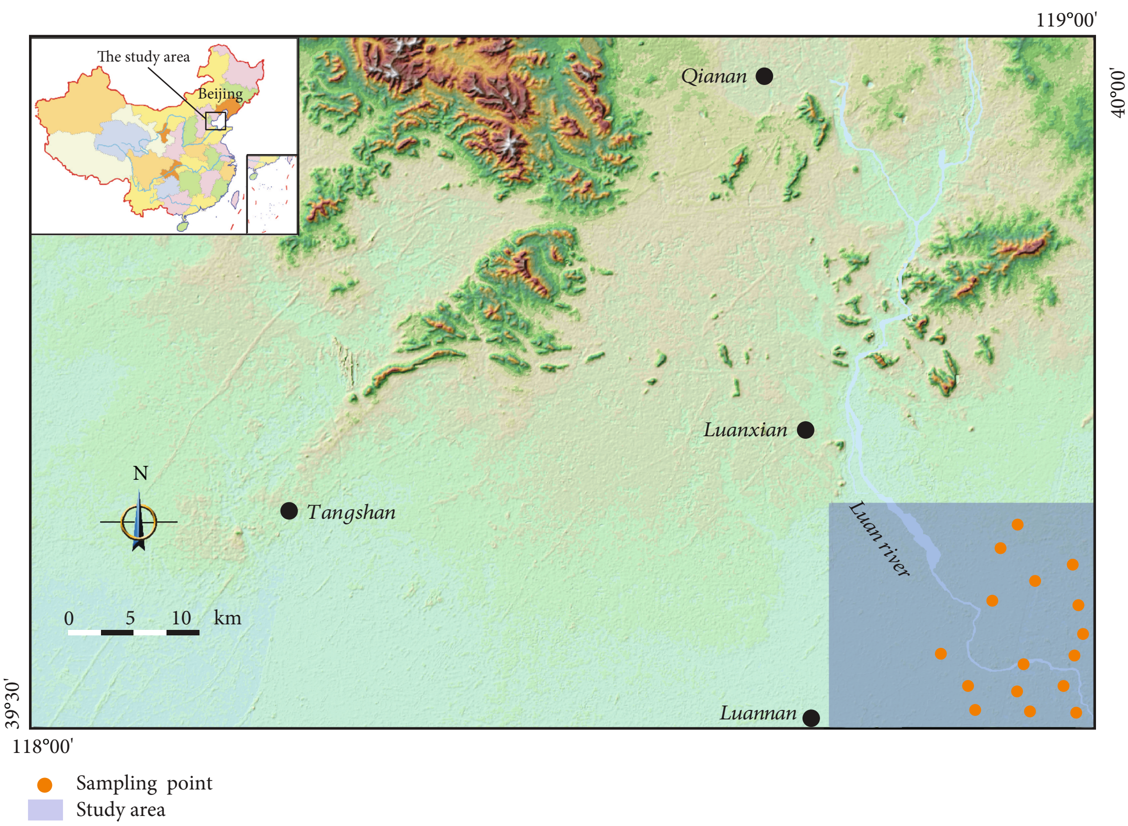

Figure 1: Location of study area and groundwater samples.

Each sampling phase and sampling site was documented with pictures and notes taken during the field activities; all of this information was compiled in a database, so the exact location of each sampling site could be determined.

3. Materials and Methods

This study was conducted to investigate the spatial and temporal distribution of heavy metals in groundwater and determine heavy metal HQs and carcinogenic risks in order to assess potential risks to public health in the Luan River catchment within the North China plain. In order to figure out aforementioned objective, the next section describes the details of this procedure and involves the following steps. The first step was to characterize the heavy metal levels in the shallow groundwater samples (144). The concentrations of 39 heavy metals were measured by ICP-MS (NexION300D), including among others, As, B, Ba, Be, Cd, Cr, Cu, Fe, Li, Mn, Mo, Ni, Sb, Se, Sn, Sr, V, and Zn. Subsequently, only three heavy metals (Cd, Cr, and Ni) that exceeded the Groundwater Quality Standard III (GB/T 14848-2017) [25], which is mainly applied to centralized drinking water sources and industrial and agricultural water, were selected for further analysis. The second step was to characterize the temporal and spatial distribution of the selected heavy metals in the study area's shallow groundwater system. The third step was to calculate the HQs and carcinogenic risks for the selected heavy metals in each of the 16 private wells to provide information regarding drinking water safety management.

Table 2: Recommended value of risk assessment model parameters via drinking water in sensitive land use.

GWCRc ($L\cdot d^{-1}$)	EFc ($d\cdot a^{-1}$)	EDc (a)	BWc (kg)	ATca (d)	GWCRa ($L\cdot d^{-1}$)	EFa ($d\cdot a^{-1}$)	Eda (a)	Bwa (kg)	ATnc (d)	WAF	ACR	AHQ
0.7	350	6	15.9	26280	1.0	350	24	56.8	2190	0.2	1.0×10^{-6}	1

Hazard Quotients and carcinogenic risks were calculated using one of the models outlined in the technical guidelines for the risk assessment of contaminated sites (HJ25.3-2014) [26] prepared by the Ministry of Environmental Protection of the People's Republic of China. As noted in the technical guidelines, the most important step is selecting the appropriate risk assessment model for the particular application. Based on the technical guidelines, the study area is considered to have sensitive land use due to the use of private wells. There are three main pathways through which human exposure to heavy metals in groundwater may occur: (a) direct ingestion, (b) inhalation through the mouth and nose, and (c) dermal absorption [19]; however, the direct ingestion of groundwater is the main pathway of exposure. Therefore, considering the sensitive land use designation of the study area and main exposure route of direct ingestion, risk assessment calculations were performed using the formulas described below from the technical guidelines for the risk assessment of contaminated sites.

The carcinogenic effect of a pollutant is estimated over the course of a lifetime, taking into consideration exposures during both childhood and adulthood. The heavy metal exposure resulting from drinking groundwater (carcinogenic effect) was calculated using the following formula [26]:

$$\text{CGWERca} = \frac{\text{GWCRc} \times \text{EFc} \times \text{EDc}}{\text{BWc} \times \text{ATca}} + \frac{\text{GWCRa} \times \text{EFa} \times \text{EDa}}{\text{BWa} \times \text{ATca}}, \quad (1)$$

where CGWERca ($L\cdot kg^{-1}\cdot d^{-1}$) refers to exposure resulting from drinking groundwater (carcinogenic effect), GWCRc ($L\cdot d^{-1}$) is the daily groundwater consumption rate for a child, EFc ($d\cdot a^{-1}$) is the exposure frequency for a child, EDc (a) is the exposure duration for a child, BWc (kg) is the average body weight of a child, ATca (d) is the average time for the carcinogenic effect, GWCRa ($L\cdot d^{-1}$) is the daily groundwater consumption rate for an adult, EFa ($d\cdot a^{-1}$) is the exposure frequency for an adult, EDa (a) is the exposure duration for an adult, and BWa (kg) is the average body weight of an adult.

In estimating the noncarcinogenic effect of a pollutant, exposures during childhood are taken into consideration. The exposure from drinking groundwater (noncarcinogenic effect) was calculated using the following formula [26]:

$$\text{CGWERnc} = \frac{\text{GWCRc} \times \text{EFc} \times \text{EDc}}{\text{BWc} \times \text{ATnc}}, \quad (2)$$

where CGWERnc ($L\cdot kg^{-1}\cdot d^{-1}$) refers to exposure resulting from drinking groundwater (noncarcinogenic effect), and ATnc (d) is the average time for the noncarcinogenic effect.

Table 3: The cancer slope factor (SFo) and reference dose (RfDo) of heavy metals via drinking water from the study area (/ indicates that no relevant data has been found).

Heavy metals	SFo ($kg\cdot d\cdot mg^{-1}$)	RfDo ($mg\cdot kg^{-1}\cdot d^{-1}$)
Cd	/	0.001
Cr	0.5	0.003
Ni	/	0.020

The carcinogenic risk to the region's population resulting from drinking groundwater contaminated with heavy metals (CRcgw) was calculated using the following formula [26]:

$$\text{CRcgw} = \text{CGWERca} \times \text{Cgw} \times \text{SFo}, \quad (3)$$

where CRcgw is the carcinogenic risk from drinking contaminated groundwater, Cgw ($mg\cdot L^{-1}$) is the contaminant concentration in the groundwater, and SFo ($kg\cdot d\cdot mg^{-1}$) is the cancer slope factor for oral intake.

To assess the health risks associated with heavy metal contamination in groundwater, HQs were calculated using the following formula [26]:

$$\text{HQcgw} = \frac{\text{CGWERnc} \times \text{Cgw}}{\text{RfDo} \times \text{WAF}}, \quad (4)$$

where HQcgw is the HQ for drinking contaminated groundwater, RfDo ($mg\cdot kg^{-1}\cdot d^{-1}$) is the oral reference dose, and WAF is the groundwater allocation factor.

The risk assessment model used the above formulas to assess the potential harm to public health in the study area. The parameter values used in formulas (1), (2), (3), and (4) are summarized in Table 2 and were taken from the technical guidelines for the risk assessment of contaminated sites.

The cancer slope factor (SFo) and reference dose (RfDo) for each heavy metal for drinking groundwater from the study area are presented in Table 3.

4. Results and Discussion

4.1. Spatial and Temporal Variations of Cd, Cr, and Ni in the Shallow Groundwater. In this study, a total of 144 groundwater samples collected over eight consecutive periods (from December 2016 to May 2017) from 16 operational private wells were subjected to chemical analysis. In order to illustrate the variations in heavy metals more intuitively, boxplots of the spatial and temporal distributions of Cd, Cr, and Ni were prepared (Figure 2). Figure 2 depicts the spatial and temporal variations of heavy metals in the shallow groundwater system from December 2016 to May 2017.

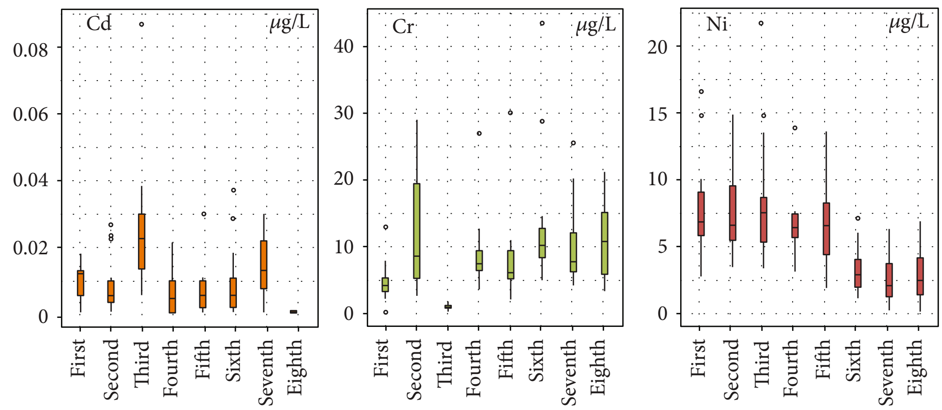

Figure 2: Spatial and temporal variations of Cd, Cr, and Ni in the shallow groundwater.

All three metals (Cd, Cr, and Ni) were detectable at every site during all eight consecutive periods. Cr had the highest concentrations, followed by Ni and Cd (Figure 2). Figure 2 shows that the select heavy metals exhibited strong variation from December 2016 to May 2017 (eight consecutive periods). The mean Cd concentrations were 0.011, 0.010, 0.026, 0.007, 0.009, 0.011, 0.016, and 0.002 μg/L for the eight consecutive periods. For Cr, the mean concentrations were 4.778, 12.106, 1.057, 8.895, 8.068, 13.379, 10.313, and 11.231 μg/L for the eight consecutive periods. The changes in Ni concentrations were sharper and more evident compared to the other heavy metals. The mean Ni concentrations were 8.073, 7.967, 8.129, 8.160, 6.891, 3.740, 3.036, and 3.232 μg/L for the eight consecutive periods.

4.2. Human Health Risk Assessment for the Study Area. In calculating both the HQs and carcinogenic risks for heavy metals in shallow groundwater in the Luan River catchment within the North China plain, the concentrations of Cd, Cr, and Ni measured during eight consecutive periods were considered. Based on formulas (1) and (2), the heavy metal exposure from drinking contaminated groundwater is 9.15 $\times 10^{-3}$ L·kg^{-1}·d^{-1} for the carcinogenic effect and 4.22 $\times 10^{-2}$ L·kg^{-1}·d^{-1} for the noncarcinogenic effect.

Today, risk assessment is one of the best approaches for investigating the potential risks of heavy metal exposure on human health, offering important information to public health decision makers for protecting consumer health [1]. Therefore, the potential health risks associated with heavy metal exposure were assessed using the data collected in this study.

The carcinogenic risk of each heavy metal was calculated separately for each consecutive period, as well as the HQs (see Table 4). The carcinogenic risk values for Cd were below the standard value of 10^{-6} [26–29] recommended by the technical guidelines for the risk assessment of contaminated sites, in all consecutive periods, except the fourth period, suggesting that there is no significant cancer risk for people living in this region (Table 4). In the fourth period, the carcinogenic risk values for Cd ranged between 10^{-6} and 10^{-4}, with an average value of 3.48×10^{-5}. The carcinogenic risk values for Ni showed the same trend in all consecutive periods with both the maximum and the minimum carcinogenic risk values exceeding 10^{-6}, implying that the groundwater is unfit for human consumption. The carcinogenic risk values for Ni ranged between 10^{-6} and 10^{-5}, with an average value of 2.81 $\times 10^{-5}$ for the eight consecutive periods. With the exception of the third period, the carcinogenic risk values for Cr showed a similar trend to Ni. The carcinogenic risk values for Cr were almost 10^{-5}, with an average value of 3.99 $\times 10^{-5}$ for the eight consecutive periods.

Hazard Quotients estimated for local residents assuming oral intake of water are summarized in Table 4. Both the minimum and the maximum HQs for Cd were less than the standard value of 1 [15, 26, 28, 30] recommended by the technical guidelines for the risk assessment of contaminated sites, in all consecutive periods, except the fourth period. The results suggest that there is no significant cancer risk to people living in this region. With respect to Ni, both the minimum and the maximum HQs exceeded 1 in all eight consecutive periods, implying that the contaminated groundwater is unfit for human consumption. The HQs for Ni ranged between 0.168 and 4.433, with an average value of 1.299 for the eight consecutive periods, which is 1.299 times greater than the standard value. The minimum and maximum HQs for Cr exceeded a value of 1 in all consecutive periods, except the third period. The HQs for Cr ranged between 0.041 and 9.161, with an average value of 2.074, which is 2.074 times greater than the standard value.

5. Conclusions

In this study, a total of 144 groundwater samples collected over eight consecutive periods (from December 2016 to May 2017) from 16 private operational wells were subjected to chemical analysis. In general, heavy metals in the Luan River catchment within the North China plain might be a threat to local residents; the HQs and carcinogenic risk values of Cd were generally in accordance with recommended standard values, and only the HQs and carcinogenic risk values of Ni and Cr exceeded the recommended standard values. Accumulation of biotoxic heavy metals in groundwater and subsequent transport into organisms may pose potential risks to human health. Therefore, the prevention and control

Table 4: The carcinogenic risks and HQs of Cd, Ni, and Cr in different periods.

Heavy metals	Period	Carcinogenic risk			Hazard Quotient		
		Max	Min	Average	Max	Min	Average
Cd	First	9.145×10^{-8}	9.145×10^{-9}	4.939×10^{-8}	4.222×10^{-3}	4.222×10^{-4}	2.280×10^{-3}
	Second	1.235×10^{-7}	9.145×10^{-9}	4.634×10^{-8}	5.699×10^{-3}	4.222×10^{-4}	2.139×10^{-3}
	Third	3.841×10^{-7}	3.201×10^{-8}	1.189×10^{-7}	1.773×10^{-2}	1.478×10^{-3}	5.488×10^{-3}
	Fourth	1.006×10^{-4}	9.145×10^{-6}	3.475×10^{-5}	4.644	0.422	1.604
	Fifth	1.372×10^{-7}	9.145×10^{-9}	4.298×10^{-8}	6.322×10^{-3}	4.222×10^{-4}	1.984×10^{-4}
	Sixth	1.692×10^{-7}	9.145×10^{-9}	4.939×10^{-8}	7.810×10^{-3}	4.222×10^{-4}	2.280×10^{-3}
	Seventh	1.372×10^{-7}	9.145×10^{-9}	7.225×10^{-8}	6.332×10^{-3}	4.222×10^{-4}	3.335×10^{-3}
	Eighth	9.145×10^{-9}	9.145×10^{-9}	9.145×10^{-9}	4.222×10^{-4}	4.222×10^{-4}	4.222×10^{-4}
Ni	First	7.408×10^{-5}	1.504×10^{-5}	3.691×10^{-5}	3.419	0.694	1.704
	Second	6.676×10^{-5}	1.802×10^{-5}	6.676×10^{-5}	3.082	0.832	1.682
	Third	6.630×10^{-5}	1.825×10^{-5}	3.717×10^{-5}	3.061	0.842	1.716
	Fourth	9.603×10^{-5}	1.660×10^{-5}	3.731×10^{-5}	4.433	0.766	1.722
	Fifth	6.127×10^{-5}	1.116×10^{-5}	3.151×10^{-5}	2.828	0.515	1.455
	Sixth	3.356×10^{-5}	8.002×10^{-6}	1.710×10^{-5}	1.549	0.369	0.789
	Seventh	3.023×10^{-5}	4.173×10^{-6}	1.388×10^{-5}	1.395	0.191	0.641
	Eighth	3.251×10^{-5}	3.640×10^{-6}	1.478×10^{-5}	1.501	0.168	0.682
Cr	First	5.944×10^{-5}	8.871×10^{-7}	2.185×10^{-5}	2.744	0.041	1.009
	Second	1.326×10^{-4}	1.225×10^{-5}	5.536×10^{-5}	6.121	0.566	2.555
	Third	8.551×10^{-6}	1.367×10^{-6}	4.834×10^{-6}	0.395	0.063	0.223
	Fourth	1.235×10^{-4}	1.687×10^{-5}	4.068×10^{-5}	5.699	0.779	1.878
	Fifth	1.372×10^{-4}	1.029×10^{-5}	3.689×10^{-5}	6.332	0.475	1.703
	Sixth	1.985×10^{-4}	2.286×10^{-5}	6.118×10^{-5}	9.161	1.055	2.824
	Seventh	1.166×10^{-4}	1.916×10^{-5}	4.716×10^{-5}	5.383	0.884	2.178
	Eighth	9.740×10^{-5}	1.550×10^{-5}	5.135×10^{-5}	4.496	0.716	2.371

of heavy metal contamination in the Luan River catchment should focus on Ni and Cr. It is, thus, required that the water sources should be properly protected from potential contamination of these harmful metals and appropriate treatment be selected for future use of water in the region.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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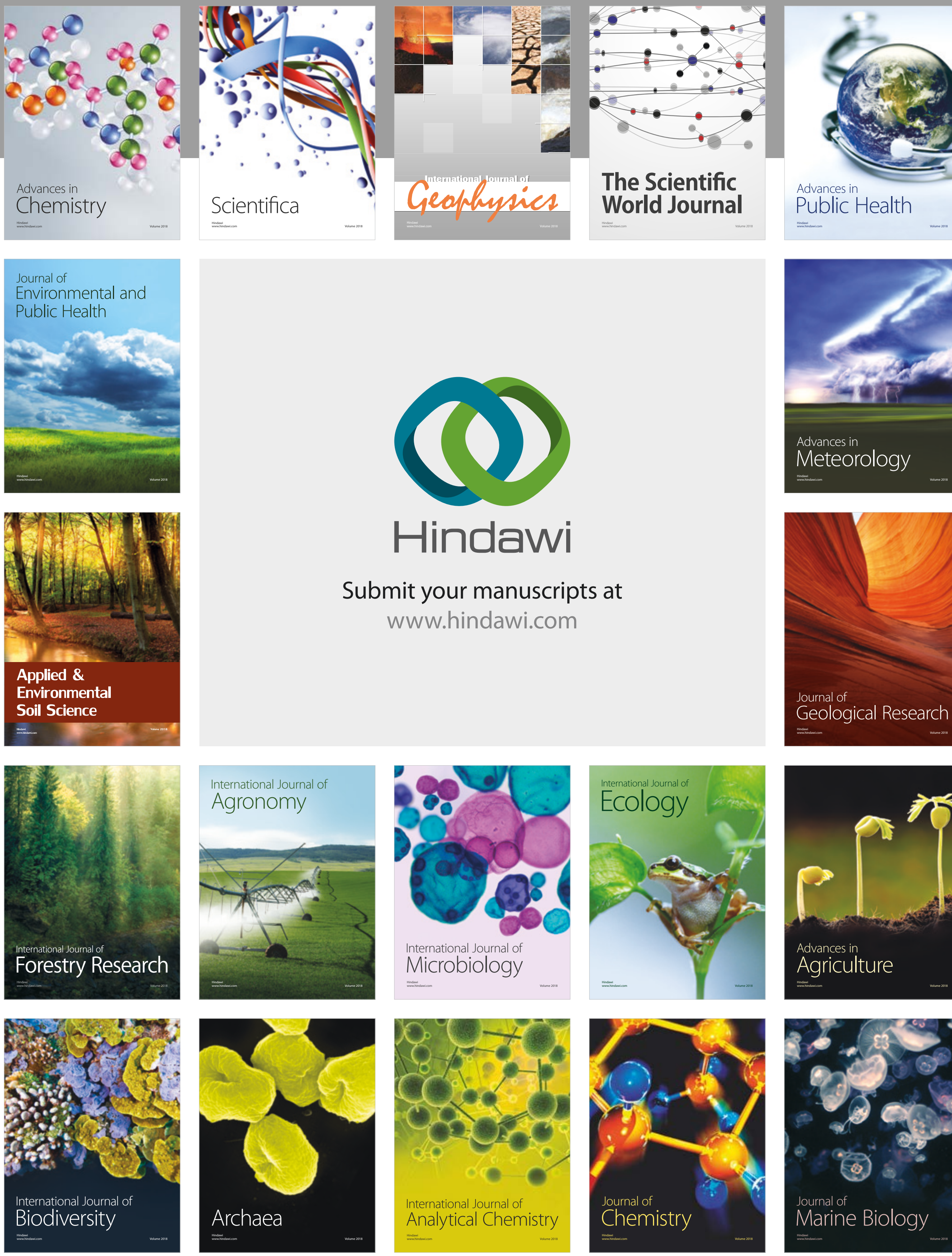
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