China Geology 7 (2024) 116−124

China Geology

Journal homepage: http://chinageology.cgs.cn
https://www.sciencedirect.com/journal/china-geology

Harmful evaluation of heavy metals from soil layer to the groundwater: Take the Jilin Hunchun Basin as an example

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ARTICLE INFO

Article history:
Received 8 April 2022
Received in revised form 11 July 2022
Accepted 16 September 2022
Available online 8 September 2023

Keywords:
Soil heavy metals
As+Hg+Cu+Pb+Zn+Ni+Cd
Environmental capacity
Groundwater
Hazard degree
Migration flux model
Agricultural geological survey engineering
Hunchun Basin
Jilin Province

ABSTRACT

The continuous enrichment of heavy metals in soils has caused potential harm to groundwater. Quantitative methods to evaluate the harm of heavy metals in soil to groundwater are lacked in previous studies. Based on the theory of groundwater circulation and solid-liquid equilibrium, a simple and easy-to-use flux model of soil heavy metals migrating to groundwater is constructed. Based on groundwater environmental capacity, an innovative method for evaluating the harm of heavy metals in soil to groundwater is proposed, which has been applied in Hunchun Basin, Jilin Province, China. The results show that the fluxes of soil heavy metals into groundwater in the study area are Zn, Cu, As, Pb, Cd, Ni, and Hg in descending order. The content of heavy metals in groundwater (As, Hg, Cu, Pb, Zn, Ni, and Cd) in most areas has not risen to the threshold of environmental capacity within 10 years. The harm levels of soil heavy metals to groundwater in the most townships soils are at the moderate level or below. This evaluation method can quantify the flux of soil heavy metals into groundwater simply and quickly, determine the residual capacity of groundwater to heavy metals, evaluate the harm level of soil heavy metals to groundwater, provide support for relevant departments to carry out environmental protection of soil and groundwater, and provide a reference to carry out similar studies for related scholars.

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1. Introduction

The enrichment process of heavy metals in soil is slow, continuous, and irreversible, which may causes soil pollution and inevitably harms the surrounding environment (Zhao KL et al., 2020; Xiao H et al., 2021; Zhang M et al., 2020; Liao JB et al., 2017; Wu GH et al., 2020). Heavy metals in the soil are leached into the groundwater system through natural and artificial activities such as precipitation and irrigation, which endangers the quality of groundwater environment (Xie F et al., 2016; Cong X et al., 2017; Li QY et al., 2021; Gao JW et al., 2021). The research on the hazard assessment of heavy metals in soil to groundwater has become an important research direction (Zhang YS et al., 2017; Wang XC et al., 2018; Bao LR et al., 2020).

In previous research, the evaluation of the harm of soil heavy metals to groundwater is very few, and the migration flux of soil heavy metals to groundwater is more. The simulation of migration characteristics of heavy metals under precipitation and acid rain conditions has been carried out by a large number of scholars (Chen GQ et al, 2010; Hu YS et al., 2020; Lü D et al., 2019; Vink JP et al., 2017; Chen ZF et al., 2014). The migration and transformation model has been used by some scholars to simulate and evaluate the migration and transformation characteristics of heavy metals in vadose zone (Lin T et al., 2019; Wan SY et al., 2020; Lin J et al., 2021). In addition to the above research directions, the evaluation method of risk screening value of heavy metal pollutants based on groundwater protection has been put forward by some scholars. The evaluation methods of risk screening value of pollutants in contaminated sites in the United States, Sweden and China were referenced. The flux model of heavy metal migration to groundwater was established, and the risk screening value of heavy metals in soil was deduced (Jiang SJ et al., 2016; Wang Y, 2013; Xu

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Literary editor: Li-qiong Jia
doi:10.31035/cg2022050
2096-5192/© 2024 China Geology Editorial Office.

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ZG, 2012; Feng Z, 2020). These methods are the basis of this study, but quantitative evaluation is lacking in these methods.

Based on the theory of water cycle and solid-liquid balance, the path of heavy metals from the soil surface into groundwater in the Chunchun Basin was deeply studied. In the basis of the characteristics of non-point source pollution, the flux model of heavy metals from soil into groundwater was established. The environmental capacity of heavy metals in groundwater was regarded as the index of heavy metal carrying capacity of groundwater, and the evaluation method of the degree of heavy metals on the soil surface to groundwater was put forward, and the hazard of heavy metals in soil to groundwater was quantitatively evaluated.

2. Materials and methods

2.1. Study area overview

Hunchun is an international cooperation demonstration zone of the Tumen River in China, which is adjacent to North Korea and Russia. Its mining industry is developing, and its economy has developed rapidly in recent years. Soil environment has become an important restriction factor for economic and social development. Hunchun Basin, located in the east of Jilin Province and the south-central part of Hunchun City, is the main population and economic distribution area of Hunchun City. It is a continental monsoon climate of offshore phase in study area, where the average annual precipitation of 618 mm (Shi XF et al., 2017). One of the main rivers in this area is Hunchun River, which flows in from northeast and out from the southwest. The other one is Tumen River, which flows from north to south along the western boundary of the study area. In the study area, some factors lead to the enrichment of heavy metals in local soil, such as developed commercial trade, frequent traffic, large-scale mining and the use chemical fertilizers and pesticides in farmland (Guo XD et al., 2022).

2.2. Sample collection and testing

Soil samples were collected from June to October 2015, with a sampling density of 1 point /km^2 and a depth of 20 cm from the surface. A total of 319 samples were collected, which were mainly arranged in different land use types such as rice fields, dry land, and other grasslands, wasteland, and woodland (Fig. 1). The soil samples were tested by Jilin Institute of Geology and Mineral Resources and digested with HNO_3-$HClO_4$-HF in the ratio of 5∶2∶3. Ni, Cd, and Pb were analyzed by plasma mass spectrometry (ICP-MS), As and Hg by atomic fluorescence spectrometry (AFS), and Cu and Zn by fluorescence spectrometry (XRF-X), and quality control was performed according to the *China Geological Survey Multipurpose Regional Geochemical Measurement Standard* (1∶250 000) (China Geological Survey, 2014).The detection limits of As, Hg, Cu, Pb, Zn, Ni, and Cd were 1 mg/kg, 0.5 mg/kg, 1 mg/kg, 0.105 mg/kg, 4 mg/kg, 0.22 mg/kg, and 0.007 mg/kg, respectively.

Groundwater samples were collected from May to September 2015, and 98 sets of groundwater samples were collected. Before collection, wells were washed and 1000 mL of water samples were taken for testing conventional anions and cations. The samples were filtered and stored in polyethylene bottles at low temperatures. All samples were sent to the Northeast Mineral Resources Supervision and Testing Center of the Ministry of Land and Resources of China for testing. The anion-cation balance of the test results was analyzed. If the error is less than 5%, the results can be used for research.

2.3. Evaluation method of the hazard of soil heavy metals to groundwater

2.3.1. Fluxes of soil heavy metals into groundwater

There is an equilibrium partitioning of pollutants in the pores of soil particles in solid, liquid, and gas phases, and the equilibrium model is Equation (1).

$$C_n = C_k \times \left(K_d + \frac{\theta_W + \theta_a \times H'}{\rho_b} \right) \tag{1}$$

Where: C_s is the concentration of soil pollutant, mg/kg; C_L is the leachate concentration of soil contaminant, mg/L; K_d is the soil solid phase - water partition coefficient, L/kg. θ_w is the soil water-filled porosity. θ_a is the soil aerated porosity. H' is Henry's constant. ρ_b is the soil bulk weight, kg/L.

For heavy metals, Henry’s constant (H') and soil aerated porosity (θ_a) can be ignored, where θ_{w} can be calculated using Equation (2).

$$\theta_w = \frac{\rho_b \times P_{ws}}{\rho_w} \tag{2}$$

ρ_w is the density of water, 1 kg/L, and P_{ws} is the soil moisture content, kg (water)/kg (soil). Then the concentration of soil leachate can be calculated by Equation (3).

$$C_L = C_s \div \left(K_d + \frac{P_{ws}}{\rho_w} \right) \tag{3}$$

K_d is the solid-liquid partition coefficient, which is the ratio of heavy metal content in the soil solid phase to the liquid phase under the condition of soil chemical equilibrium, and its value is influenced by a variety of factors. Soil solid-liquid partition coefficient is greatly influenced by pH, redox environment, organic matter, and other factors. Global scholars have done a lot of in-depth research, the calculation method of K_d for each element draws on previous research results, the calculation method is shown in Table 1.

Based on the principle of heavy metal solid-liquid equilibrium, the precipitation infiltration pathway and irrigation infiltration pathway models of soil heavy metals into groundwater were constructed, and the flux Q_g of soil surface heavy metals into groundwater could be calculated by Equation (4).

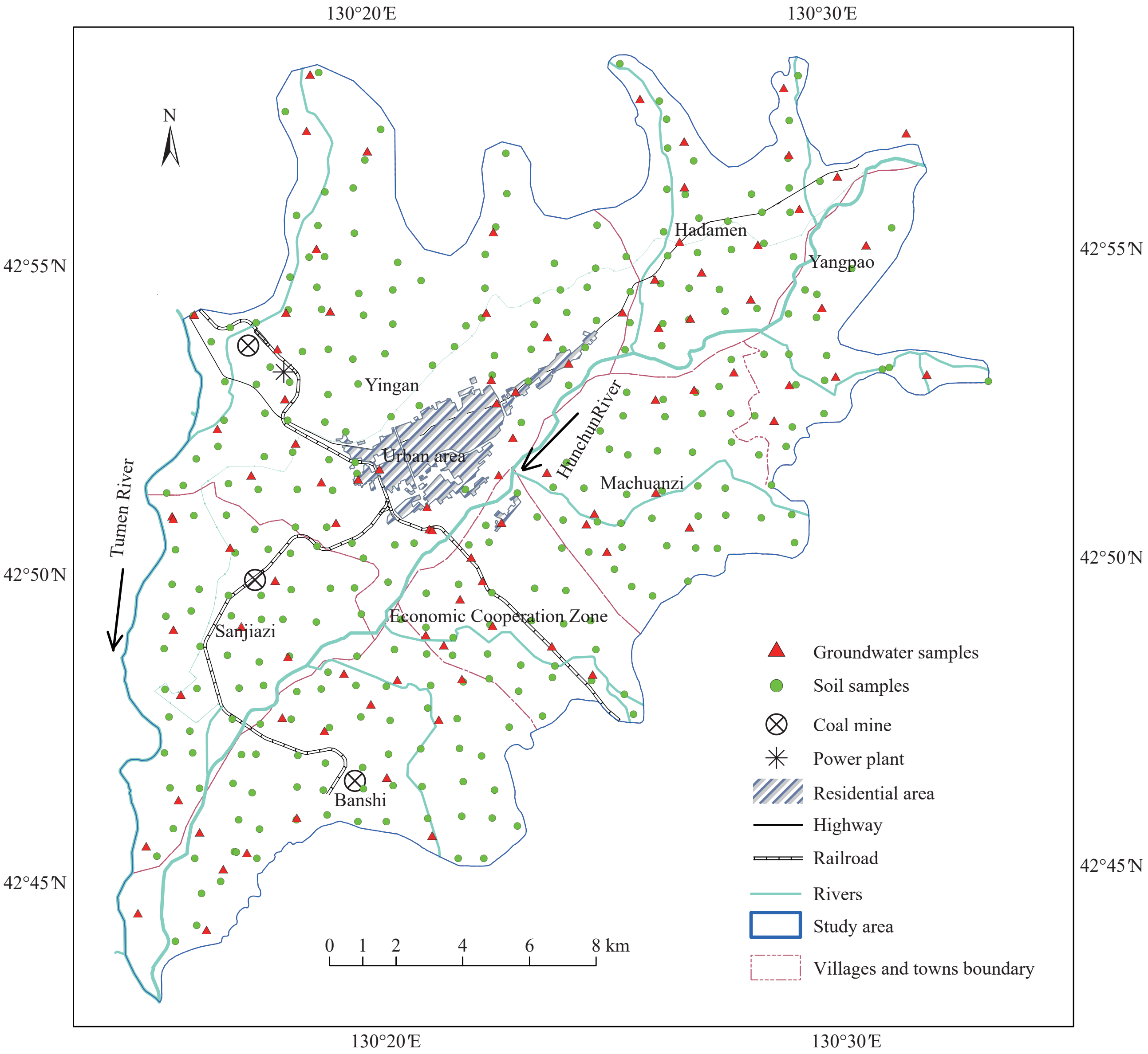

Fig. 1. Distribution map of soil samples and water samples in Hunchun Basin.

Table 1. List of prediction models for solid-water partition coefficients of each element.

Elements	K_d prediction model	Reference
As	Lgk_d=0.41lg (total)+0.72lg (AlFeox)−0.40	Groenenberg JE et al., 2012
Hg	Dry fields, lgK_d=0.021lg (Slit)+2.70 Paddy fields, lgK_d=1.13lg (S)+3.17	Wang XC et al, 2018
Cu	Lgk_d=0.21pH +0.51lg (SOM)+1.75	Sauve S et al, 2000
Pb	Lgk_d=0.37pH +0.44lg (total)+1.19	Sauve S et al, 2000
Zn	Lgk_d=0.60pH +0.21lg (total)−1.34	Sauve S et al, 2000
Ni	Lgk_d=1.02pH +0.80lg (SOM)−4.16	Sauve S et al, 2000
Cd	Lgk_d=0.48pH+0.82lg (SOM)−0.65	Sauve S et al, 2000

Notes: SOM is organic carbon content in %C, the total is total elemental content in mg/kg; AlFeox is iron and aluminum oxide in % (iron and aluminum oxide/soil). Slit: Soil clay grain content in %, S is total soil sulfur content in g/kg.

$$Q_g = 10^{-1}\times \alpha \times F + \beta \times Q_{iw}) \times C_s \div \left(K_d + \frac{P_{ws}}{\rho_w}\right) \tag{4}$$

Where: Q_g is the amount of soil heavy metals in groundwater (g/a); P_r is the annual precipitation infiltration recharge (10^4 m^3/a); α is *the* precipitation infiltration recharge coefficient, dimensionless; P is the annual precipitation (mm); F is the area (km^2); Q_r annual field irrigation infiltration recharge (10^4 m^3/a); β is the field irrigation infiltration recharge coefficient, dimensionless; Q_{iw} annual irrigation water (10^4 m^3). Other quantities are shown in Equation (1) and Equation (2).

2.3.2. Groundwater environmental capacity

The environmental capacity of medium heavy metals in groundwater reflects that the groundwater can hold more heavy metals under a certain limit of heavy metal content, and reflects the pollutant-carrying capacity of groundwater. The hazard of soil heavy metals to groundwater can be measured by the number of heavy metals that the groundwater environment can continue to hold. The capacity of heavy metals in groundwater is obtained by the product of the average annual reserves of groundwater and the average annual residual content of heavy metals in groundwater.

The environmental capacity of groundwater can then be based on the quality standard limits of heavy metals class III in GB 14848-2017 Groundwater Quality Standard as the capacity limits of pollutants that can be consumed in groundwater (General Administration of Quality Supervision, Inspection and Quarantine of the People’s Republic of China, 2017). Then the groundwater environmental capacity can be calculated by Equation (5).

$$Q_d = (C - C_g)\times \mu \times H_w \times F \tag{5}$$

Where: Q_d is the environmental capacity of heavy metals in groundwater (g); C_g is the content of heavy metals in groundwater (mg/L); C' is the pollution limit value of heavy metals in groundwater (mg/L), referring to the Ⅲ quality standard in GB 14848-2017 Groundwater Quality Standard; μ is the aquifer feed degree, dimensionless; H_w is the thickness of underground aquifer (m); F is the area (m^2).

2.3.3. Hazard degree of soil heavy metals to groundwater

The flux of regional soil heavy metals into groundwater reflects the number of heavy metal pollutants entering the groundwater system every year, and the ratio of the number of heavy metals entering the groundwater system to the environmental capacity of heavy metals in groundwater can reflect the hazard degree (Dg) of soil heavy metals to groundwater. The calculation formula is shown in Equation (6). As the pollution of heavy metals to groundwater is a slow-changing hazard, the degree of hazard can be evaluated from different periods, and the degree of hazard can be judged by reaching 50 times the environmental capacity and environmental capacity in the next 25 years, 10 years, 5 years and 1 year, respectively. See Table 2 for evaluation criteria.

$$D_g = Q_g / Q_d \tag{6}$$

3. Results and discussion

3.1. Fluxes of heavy metals into groundwater from soils in the Hunchun Basin

Precipitation and farmland irrigation are the migration routes of heavy metals from soil to groundwater in Hunchun Basin (Moon JW et al., 2000; Guo XD et al., 2018). Precipitation data was obtained from local meteorological departments, multi-year average precipitation was collected from a total of eight stations, and Tyson polygons were used for regional precipitation distribution. Precipitation infiltration coefficients were obtained according to local hydrogeological conditions and pumping tests. The irrigation volume of agricultural land was calculated based on the irrigated area of agricultural land and irrigation quota. Soil solid phase-water pollutant distribution coefficient K_d was obtained using the equation in Table 1.

Table 2. Hazard evaluation criteria of soil heavy metals to groundwater.

Hazard degree (D_g)	Hazard level
>50 & <0	Extremely serious
1–50	Severity
0.2–1.0	Moderate
0.1–0.2	Light
0.04–0.10	Alert
0–0.04	No hazard

Since the water field area is mainly infiltrated under irrigation water during the irrigation period, and there is no infiltration of atmospheric precipitation, it is necessary to deduct the infiltration of precipitation from May to August in the water field area in the calculation. Soil water content P_{ws} was set to 0.1 according to the recommended value in the *Technical Guidelines for Risk Assessment of Contaminated Sites*.

Each quantity in Equation (4) was made into an ArcGIS raster file with a raster size of 100 m×100 m, and the distribution of the amount of soil heavy metals entering groundwater in the study area was calculated. The results of the distribution were counted according to each township, and the results are shown in Table 3 and Fig. 2. The fluxes of heavy metals into groundwater in the study area were Zn, Cu, As, Pb, Cd, Ni, and Hg in descending order.

3.2. Environmental capacity of heavy metals in groundwater of Hunchun Basin

The groundwater in the study area is mainly Quaternary phreatic water. The thickness of the aquifer gradually increases from the upstream to the downstream of the Hunchun River along the northeast to the southwest, from 4 m to 7 m in the upstream area and 10 m to 12 m in the downstream area. The north and south edges of the study area are close to the hill area, and the aquifer thickness becomes thinner, the grain is fine, the stable aquifer is lacking, and the specific yield is small, ranging from 0.01 to 0.07. The central valley area is an alluvial proluvial sand gravel aquifer with coarse particles and a large water yield, between 0.10 and 0.24.

The content of each heavy metal in groundwater was obtained from the analysis of groundwater samples collected

Table 3. Fluxes of soil heavy metals into groundwater in villages and towns (kg/a).

Township	As	Hg	Cu	Pb	Zn	Ni	Cd
Banshi	114.96	1.07	136.27	18.84	1375.88	2.35	5.17
Sanjizi	98.02	0.78	133.36	18.07	1440.23	2.29	6.34
Economic Cooperation Zone	59.29	0.61	73.56	11.16	736.82	1.32	2.15
Machuanzi	103.66	1.16	114.23	19.20	1405.70	2.31	3.15
Yangpao	58.28	0.36	76.79	10.22	695.80	1.35	2.84
Yingan	179.96	1.52	240.91	32.13	2178.65	3.94	7.37
Hadamen	68.78	0.41	102.35	14.27	1011.68	1.83	4.08
Total	682.94	5.91	877.49	123.89	8844.75	15.40	31.09

in the Hunchun Basin in 2014, and the results are shown in Table 4.

The environmental capacity of each heavy metal in groundwater in the study area was calculated according to Equation (5), based on the aquifer thickness, specific yield, and heavy metal content in groundwater. The environmental capacity of heavy metals in groundwater in each township and the statistics were carried out, which are shown in Table 5 and Fig. 3. The zoning trend of the environmental capacity of heavy metals in groundwater is consistent with the groundwater storage, which is small in the upstream, large in the downstream, small in the north and south, and large in the middle. The environmental capacity of different heavy metals is not consistent, Cu and Zn have the larger environmental capacity, followed by Ni, As, Pb, and Cd, and Hg has the smallest environmental capacity.

3.3. Hazardous degree of heavy metals to groundwater in soils of Hunchun Basin

The hazard level of soil heavy metals in groundwater in the study area was calculated according to Equation (6) with ArcGIS raster calculation, and the zoning map of the mean hazard level of each heavy metal is shown in Fig. 4. No hazard or alert level occupies a large area in the study area,

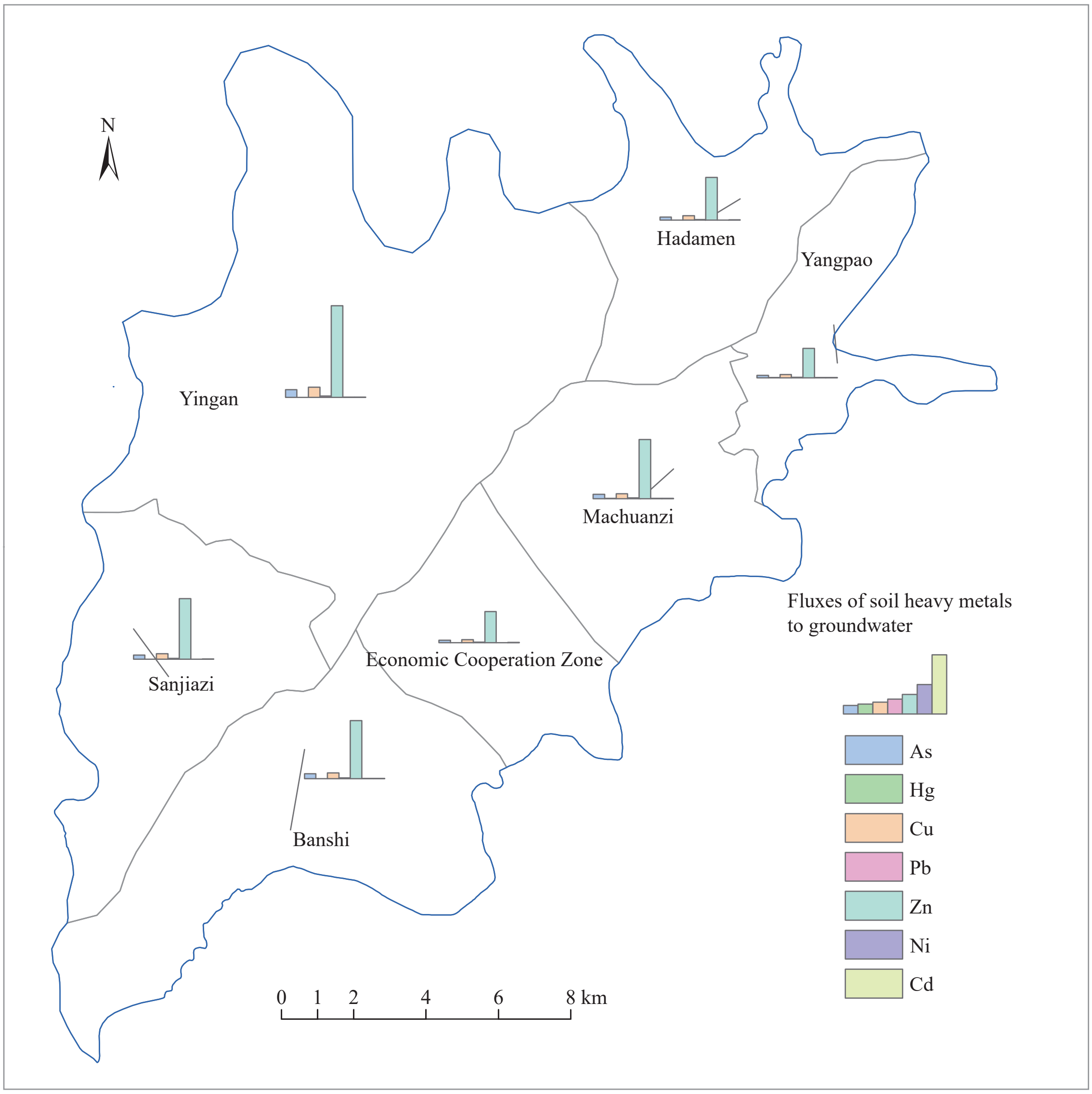

Fig. 2. Flux map of soil heavy metals into groundwater in villages and towns.

Table 4. Content of heavy metals in groundwater in the study area (mg/L).

	Cu	Pb	Zn	As	Hg	Cd	Ni
Minimum value	0.00060	0.00040	0.0036	0.0014	0.0000000	0.000000	0.0006
Maximum value	0.00910	0.01880	0.4658	0.0094	0.0000357	0.000771	0.0458
Average value	0.00483	0.00244	0.0239	0.0026	0.0000230	0.000084	0.0109
GB level III	1.00	0.01	1.00	0.01	0.001	0.005	0.02

Table 5. Environmental capacity of heavy metals in groundwater in the study area (kg).

Township	As	Hg	Cu	Pb	Zn	Ni	Cd
Banshi	692.77	92.58	94223.22	545.13	92380.17	706.93	466.76
Sanjizi	782.23	100.41	102353.49	702.37	100951.68	1460.23	503.68
Economic Cooperation Zone	126.96	16.74	17014.08	131.77	15952.48	38.10	84.17
Machuanzi	230.02	29.93	30468.46	238.12	29753.14	316.12	150.41
Yangpao	173.52	21.76	22230.00	190.63	22081.60	267.84	109.89
Yingan	745.06	101.97	103863.29	809.86	102593.92	1082.58	513.06
Hadamen	282.79	36.57	37362.07	342.22	37177.10	334.56	184.90

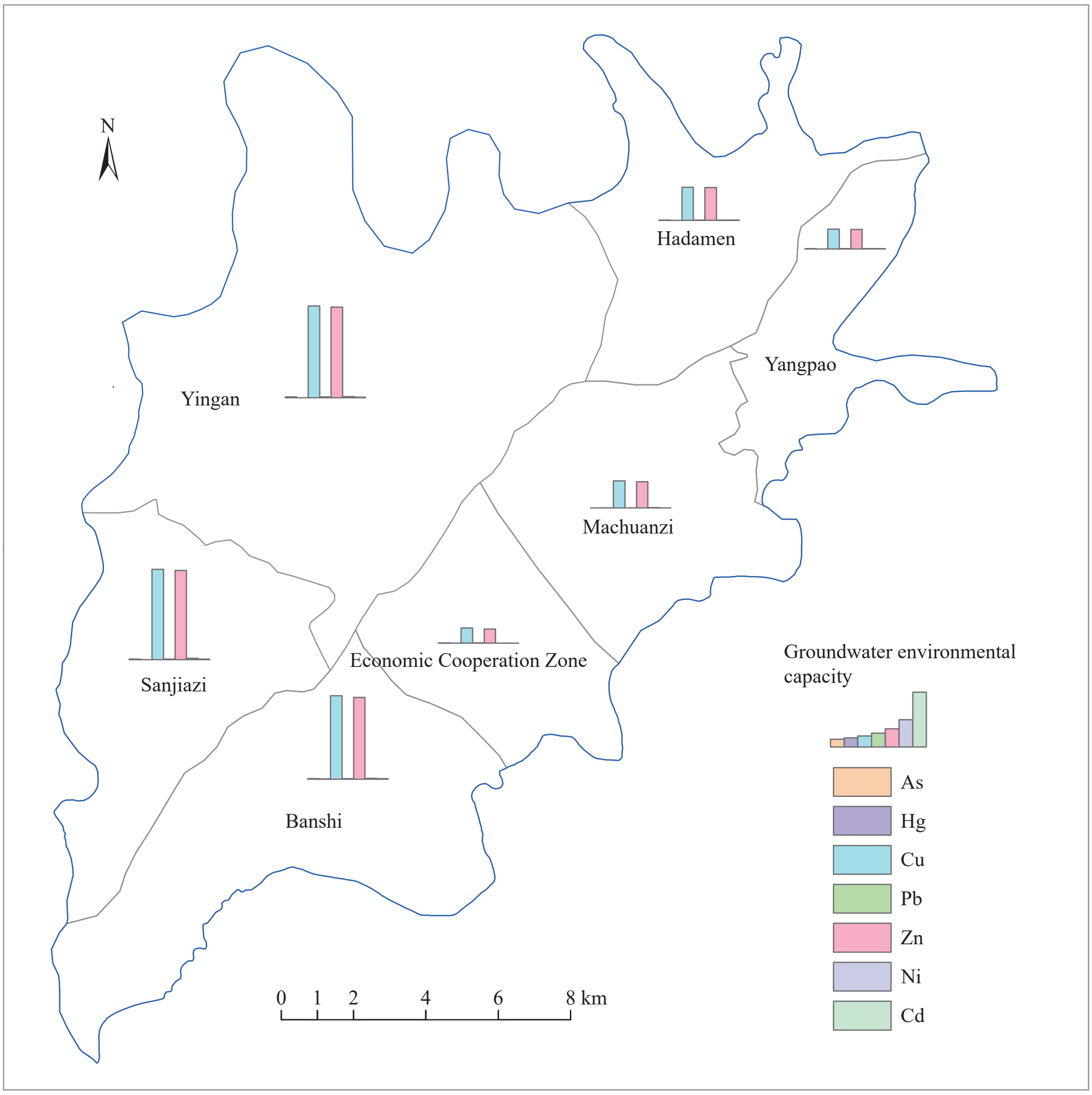

Fig. 3. Distribution map of groundwater environmental capacity in villages and towns.

and heavy metals enter the groundwater system from the surface, 39% of the area will not reach the environmental capacity limit within 25 years, 41% of the area will not reach the environmental capacity limit within ten years, 16% of the area will reach the limit within five to ten years, and another 4% of the area will reach the limit within five years.

3.4. Analysis of evaluation results

The hazard degree of soil heavy metals to groundwater is affected by many factors, such as the content of heavy metals in soil, the mobility of heavy metals, the quality of groundwater and so on. The hazard level of soil heavy metals into groundwater in the study area is shown in Table 6, which shows that the hazard level of Ni in the Economic Cooperation Area is Extremely serious, and its flux of Ni into groundwater is 1.32 kg/(a·km^2), which is no sudden change compared with other areas. However, the groundwater environmental capacity in this area is small, and some area are negative. The hazard level is negative , indicating that the

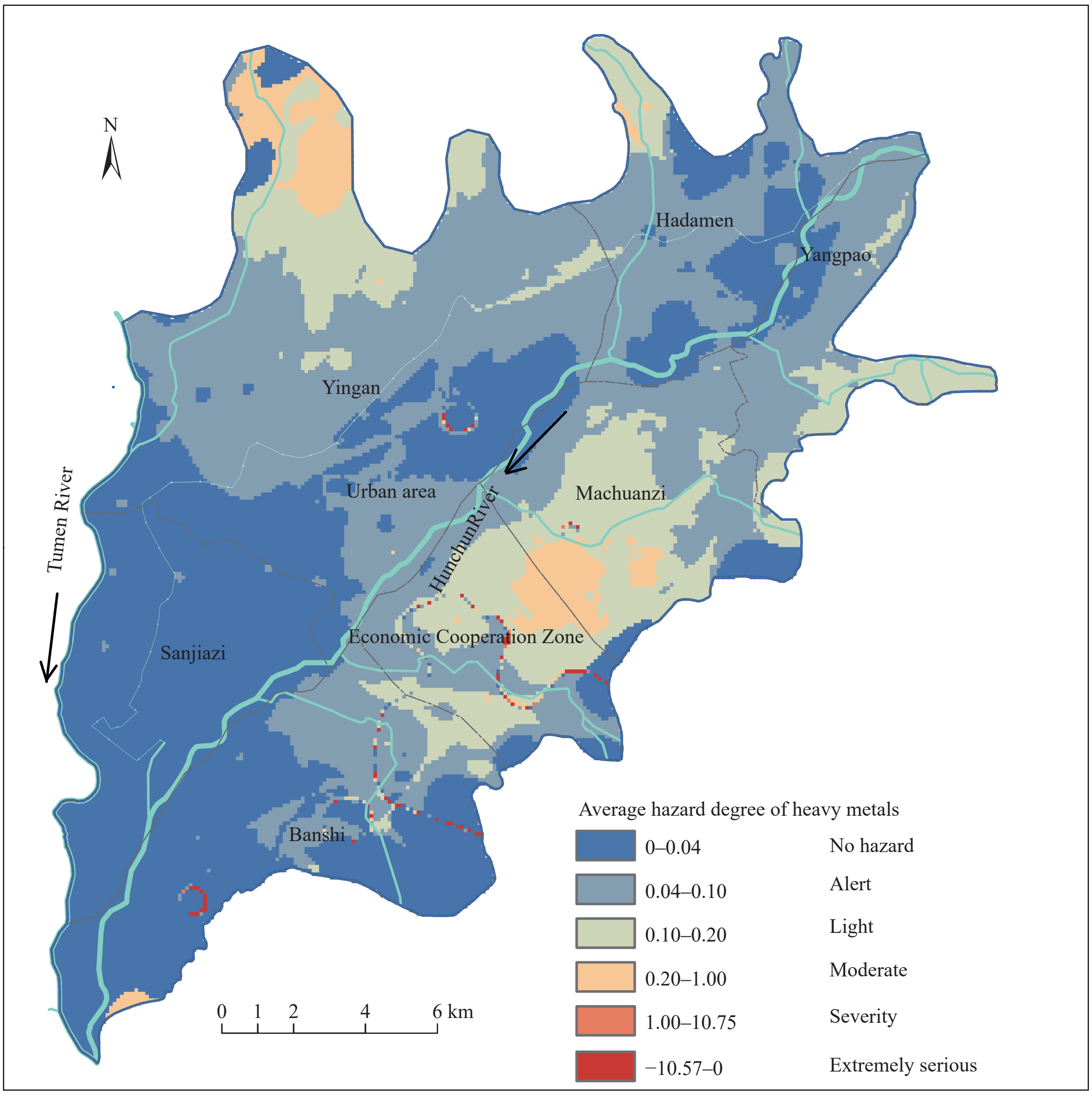

Fig. 4. Map of hazard degree of soil heavy metals to groundwater in the study area.

Table 6. List of the hazard degree of soil heavy metals to groundwater in the study area.

Township	Hazard level							
	As	Cd	Cu	Hg	Ni	Pb	Zn	Overall
Banshi	Moderate	None	None	None	None	Alert	None	Moderate
Sanjizi	Light	None	None	None	None	None	None	Light
Economic Cooperation Zone	Moderate	None	None	Alert	Extremely serious	Alert	Alert	Extremely serious
Machuanzi	Moderate	None	None	Alert	None	Alert	Alert	Moderate
Yangpao	Moderate	None	None	None	None	Alert	None	Moderate
Yingan	Moderate	None	None	None	None	Alert	None	Moderate
Hadamen	Moderate	None	None	None	None	Alert	None	Moderate

groundwater environment in this area has been seriously endangered and cannot accept the external pollution load. An extremely serious hazard level is appropriate. Except Sanjiazi Township, all the other towns and villages are moderate level, mainly because of the high hazard degree of As. The flux of As into groundwater is between 58.28 kg/(a·km^2) and 179.96 kg/(a·km^2), while the groundwater environmental capacity of As is between 1.27 kg/km^2 and 241.73 kg/km^2, resulting in a hazard degree of mostly moderate or above, indicating that the local groundwater environmental capacity is small and insufficient to support or accommodate the current heavy metal input load.

The hazard degree of soil heavy metals to groundwater reflects the quality of heavy metals in soil from the angle of protecting groundwater quality. For the study area, the high degree in the Economic Cooperation Zone reflects that the environmental capacity of groundwater in some areas of this area is small, the flux of soil heavy metals into groundwater is large, and the groundwater is greatly endangered by heavy metals, so the water quality will continue to deteriorate. Therefore, it is necessary to take engineering measures to reduce the content of heavy metals in soil and cut off the passage of heavy metals into groundwater. Other towns and villages have “moderate ” and “light ” hazards, so the monitoring of soil heavy metals and groundwater environmental quality can be carried out regularly, and prevention and control measures can be taken in time.

4. Conclusions

Based on the solid-liquid equilibrium theory of metal in soil and groundwater circulation theory, the flux evaluation model of soil heavy metals into groundwater was constructed, and the evaluation method of the environmental capacity of groundwater heavy metals was proposed, and the evaluation method of the environmental hazard of soil heavy metals to groundwater was constructed and applied in the Hunchun Basin. The following conclusions were drawn.

(i) Based on precipitation and irrigation infiltration, combined with the solid-liquid equilibrium model of soil heavy metals, an evaluation method for the flux of soil heavy metals into groundwater was established and applied in Hunchun Basin. The results showed that the fluxes of soil heavy metals into groundwater in the study area were Zn, Cu, As, Pb, Cd, Ni, and Hg in turn.

(ii) The environmental capacity evaluation method of heavy metals in groundwater was proposed and calculated in Hunchun Basin. The results showed that the environmental capacity of heavy metals in groundwater in the Hunchun Basin is larger for Cu and Zn, followed by Ni, As, Pb, and Cd, and the smallest for Hg. Among them, Pb and Ni have the negative environmental capacity in some areas.

(iii) Based on the evaluation methods of soil heavy metals into groundwater flux and groundwater environmental capacity, the evaluation method of the hazard degree of soil heavy metals to groundwater was put forward and applied in Hunchun Basin. The results showed that the heavy metals content in most areas of Hunchun Basin did not reach the environmental capacity limit in ten years. Except for the Economic Cooperation Zone, which was at an extremely serious level, the other administrative regions were at a moderate and below level. This method can quantify the hazard status of soil heavy metals to groundwater, and provide support for the environmental protection of soil and groundwater.

CRediT authorship contribution statement

Xiao-Dong Guo and Chuan-Yu Qin conceived the presented idea. Xu-Fei Shi and Zhi-Qiang Zhang participated in the collection of water samples and soil samples. Xiao-Dong Guo, Qiang Liu, and Hui-rong Zhang discussed the results. All authors contributed to the final manuscript.

Declaration of competing interest

The authors declare no conflicts of interest.

Acknowledgement

This research was jointly supported by the project of China Geology Survey (12120115032801, DD20190340).

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