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Evaluation and Source Analysis of Heavy Metal Pollution in Grassland Soils under Different Management Modes in Altay, Xinjiang

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Abstract: In order to study the characteristics and sources of heavy metal pollution within different grassland soil categories in Altay, 147 soil samples were collected in Fuyun County according to grassland type, and the contents of seven heavy metals (Cu, Pb, Cr, Zn, As, Ni, and Cd) were determined. The heavy metal pollution in the soil was evaluated using the Nemerow pollution and geological accumulation index methods. The absolute principal component score–multiple linear regression model (APCS-MLR) model was used to analyze the sources of heavy metals and the contribution rate of the pollution sources. The results showed that (1) the heavy metal content of 0~10 cm soil in the mining area was generally higher than the remaining two layers. Natural grassland was the most seriously contaminated, and the order of its contamination was Zn > Pb > Ni > Cd > Cu > Cr > As. (2) The evaluation results of the comprehensive pollution index showed that Ni and Cd were the main pollutants in five grassland types, and the evaluation results of the geological accumulation index method showed that natural grasslands were more polluted. (3) Three pollution sources were obtained according to the analysis using the APCS-MLR model, including natural and mining activity sources (32.6%), industrial coal combustion and sewage sources (24.7%), and an unknown source (42.7%). The results of the study can provide a basis for pollution control and ecological protection in the grasslands of Altay.

Keywords: grassland; heavy metals; pollution evaluation; source analysis

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Citation: Hu, Q.; Li, J.; Wang, Y.; Huang, P.; He, X. Evaluation and Source Analysis of Heavy Metal Pollution in Grassland Soils under Different Management Modes in Altay, Xinjiang. *Agronomy* **2023**, *13*, 2621. https://doi.org/10.3390/agronomy13102621

Received: 20 September 2023
Revised: 13 October 2023
Accepted: 13 October 2023
Published: 16 October 2023

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1. Introduction

Soil is among the most critical sources upon which human beings depend on for survival and development and has an irreplaceable role in ecological function [1–3]. With societal development, the soil environment is increasingly affected by human activities, and heavy metal pollution in soil has become a global problem [4,5]. Soil heavy metal pollution mainly originates from natural and human activities, with natural sources including pollution of the environment caused by soil rocks during formation. On the other hand, human activities encompass a wide variety of sources, mainly including industrial pollution, such as metal smelting and mining [6–8]; pollution sources from transportation and transport processes [9]; and agricultural pollution sources from fertilizer use and sewage discharge [10]. Soil heavy metal pollution has the characteristics of concealment and accumulation [11], which can reduce soil quality [12], have a direct impact on agriculture and animal husbandry [13], and even endanger human health [14]. Therefore, the accurate identification and quantitative analysis of sources of heavy metal pollution are prerequisites for better understanding the heavy metal pollution situation in the soil more accurately and formulating more effective restoration and protection measures.

Grassland is the most common base for the development of livestock farming and plays an important role in the ecological environment [15]. Due to the influence of anthropogenic activities, the soil environment of grassland areas has been damaged, and the accumulation of heavy metals is one of the reasons for the destruction of the soil environmental quality [16,17]; therefore, it is imperative to protect grassland soil. At present, scholars around the world have carried out research on heavy metal pollution in grasslands, finding that the soils of the northeastern Tibetan Plateau are heavily polluted with Pb, Cd, and Hg due to anthropogenic activities [18]. Anaman et al. studied heavy metal pollution in different land types, and grassland soils were more polluted with As, Cd, Pb and Cu than agricultural and forest land areas [19]. Cheng et al. studied the heavy metal content of grassland soils in Inner Mongolia, and Cu and Zn exceeded the local background values [20]. There may be a trend of increasing heavy metal concentrations in grassland soils in southern New Zealand due to increasing land use intensity [21]. For the source analysis of heavy metals in grassland soils, Gao et al. analyzed the source of heavy metals in grassland soils in Inner Mongolia by employing principal component analysis and cluster analysis [22], and Wang et al. used correlation analysis and principal component analysis for investigations of a similar nature [23]. Xu et al. used the positive definite matrix factor decomposition (PMF) model to quantitatively trace the source of soil pollution in the Gansu grassland. Traffic emissions and industrial activities were the main sources of soil heavy metals [24]. Xu et al. used the APCS-MLR model to study soil pollution in Ningxia grasslands, with industrial activities, agricultural use, and traffic as the main sources [25]. The rational assessment of soil environmental quality and the quantitative traceability analysis of heavy metal pollution sources are important for grassland pollution management.

Located in the northern part of Xinjiang, Altay is rich in mineral resources and is an important area of strategic resource reserve for non-ferrous metals in China. Animal husbandry and industrial product ores and cement production in Fuyun County have had a serious impact on the local environment. The mining industry has led to several environmental problems, such as landscape destruction and soil and water pollution, which are increasingly expanding with ecological problems and even threatening the health of residents. In this paper, different grassland types in Fuyun County were taken as the research objects, and the contents of seven heavy metals, i.e., Cu, Pb, Cr, Zn, As, Ni and Cd, were analyzed. The soil quality and pollution status were evaluated using the Nemerow pollution and geological accumulation index methods, and the sources of heavy metals in the soil were analyzed based on the APCS-MLR model to carry out a reasonable assessment of the grassland soil environment in this area and provide a basis for grassland management and pollution prevention and control in Altay.

2. Materials and Methods

2.1. Overview of the Study Area

Fuyun County is located in the northern part of Xinjiang and the southern foot of the Altay Mountain (88°10′–91°31′ E; 45°00′–48°03′ N), bordering Qinghe County in the east, Fuhai County in the west, the Junghar Basin and Changji Hui Autonomous Prefecture in the south, and Mongolia in the north. The study area is schematically shown in Figure 1. The county belongs to the continental cold-temperate arid climate and is rich in mineral resources, with copper, lead, and zinc ranking among the top in Xinjiang, but mining has become a critical problem that damages the ecological environment of Fuyun County. There are eight main types of soil, including meadow soil, wind-sand soil, and brown calcium soil. Brown calcium soil has the largest area, accounting for 43.12% of the county's arable land, and the available natural pastures amount to 4×10^6 ha.

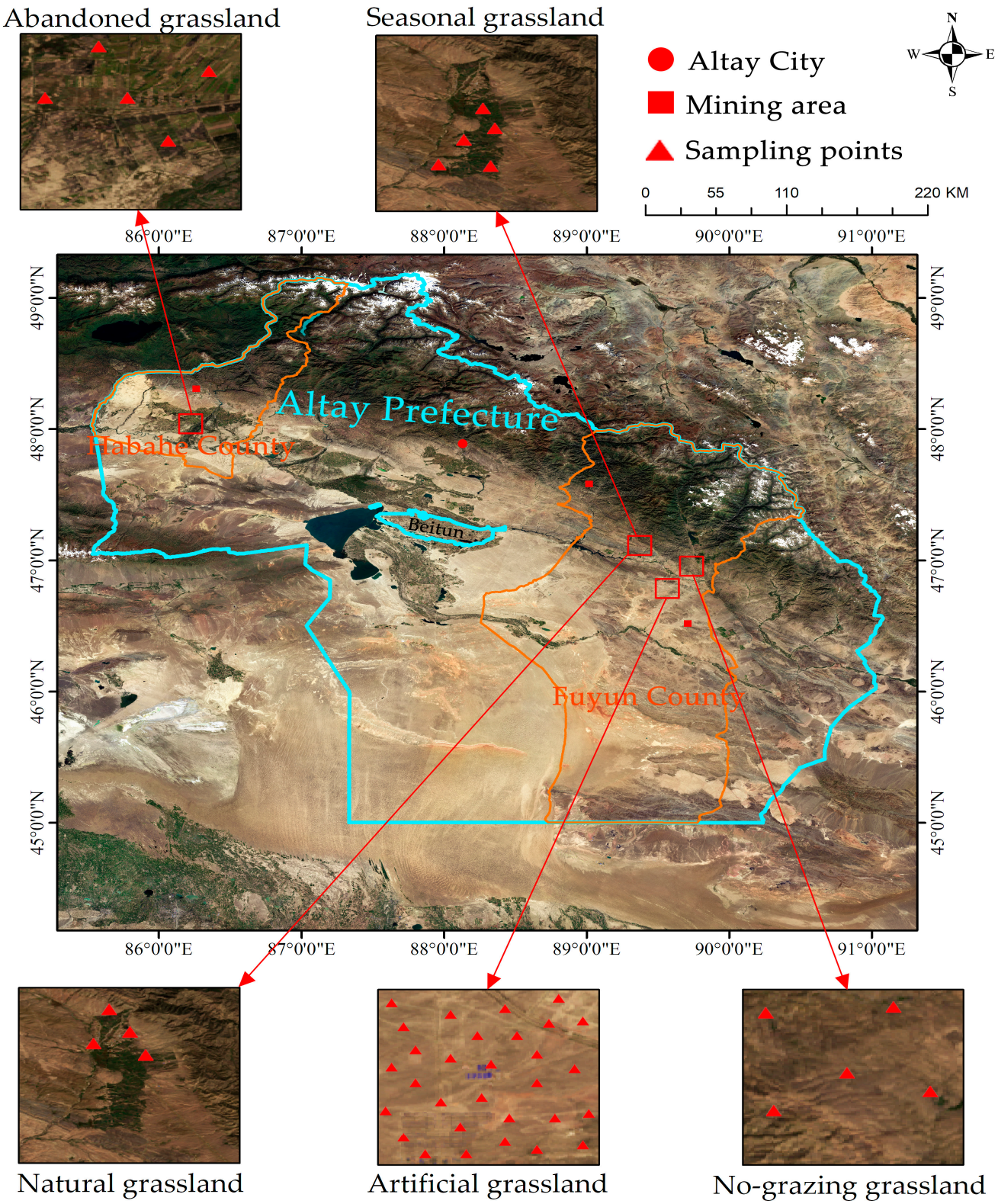


Figure 1. Schematic of the study area.

2.2. Sample Collection and Processing

Fuyun County was selected as a typical study area, and the soil samples were taken at 0~10, 10~30 and 30~50 cm depths along the diagonal line, according to the equidistant method. A total of 30 sample squares of 1 m × 1 m were set as an artificial grassland,

and 90 soil samples were taken by digging a profile in the center of each sample square. In addition, four types of grasslands, namely no-grazing grassland, seasonal grassland, abandoned grassland and natural grassland, were selected. GPS was used to locate them, and 1 m × 1 m sample squares were set up with sample ropes. Following that, five sample squares were set up for each type of grassland, except for natural grassland, which had four sample squares set up, and each sample square was divided into three soil layers to make up 57 soil samples. The heavy metal content of the samples was determined using a portable X-ray fluorescence spectrometer (Thermo scientific Niton_ XL3t_GOLDD analyzer, Niton, MA, USA).

2.3. Evaluation Methodology

2.3.1. Nemerow Pollution and Geological Accumulation Index Methods

Using the background values of the Xinjiang soil as the basis for evaluation, the Nemerow pollution [26] and the geological accumulation index methods [27] were used to evaluate the level of heavy metal pollution in the soil of Altai grassland, and the pollution levels were classified [28] as shown in (Table 1). The formula is as follows:

$$P_i = C_i / S_i \quad (1)$$

$$P_N = \sqrt{\frac{(C_i/S_i)^2_{\max} + (C_i/S_i)^2_{ave}}{2}} \quad (2)$$

where P_i is the pollution index of soil heavy metal i; C_i is the measured value of soil heavy metal content i; S_i is the evaluation background value of soil heavy metal i selected as desired; $(C_i/S_i)_{max}$ and $(C_i/S_i)_{ave}$ represent the maximum and average values of the pollution index of soil heavy metal i, respectively; and P_N is the comprehensive pollution index.

$$I_{geo} = \log_2[C_n / K \bullet B_n] \quad (3)$$

where I_{geo} is the geoaccumulation index, C_n is the measured value of soil heavy metal element content in the study area, B_n is the background value of soil heavy metal elements in Xinjiang, and K is the coefficient of variation of the background value caused by diagenesis (generally going to K = 1.5).

Table 1. Level classification standards of the soil heavy metal pollution.

Nemerow Index				Geological Accumulation Index		
P_i	**Pollution Level**	**P_N**	**Pollution Level**	**I_{geo}**	**Grading**	**Pollution Level**
$P_i \leq 1$	Cleaning	$P_N \leq 0.7$	Cleaning	<0	0	Non-polluting
$1 < P_i \leq 2$	Light pollution	$0.7 < P_N \leq 1$	Alert Line	0–1	1	Light pollution
$2 < P_i \leq 3$	Moderate pollution	$1 < P_N \leq 2$	Light pollution	1–2	2	Moderate pollution
$P_i > 3$	Severe pollution	$2 < P_N \leq 3$	Moderate pollution	2–3	3	Moderate pollution
		$P_N > 3$	Severe pollution	3–4	4	Heavy pollution
				4–5	5	Heavy pollution
				>5	6	Severe pollution

2.3.2. Absolute Principal Component Score–Multiple Linear Regression Model (APCS-MLR)

The APCS-MLR model takes pollutants as the research object and can be used without the requirement of knowing the number of pollutants first. The realization conditions are simple, and the source analysis results are reliable and accurate [29], making it suitable for the study of soil heavy metal pollution in the Altay area. The absolute principal component score–multiple linear regression model [30] was applied to quantitatively identify the main pollution sources and contributions of seven heavy metals in Altay grassland soils.

3. Results

3.1. Concentrations of Heavy Metal in Soil

The contents of seven heavy metals in different layers of grassland soil under different management modes in Altay are shown in Table 2. From this table, it can be seen that the contents of five kinds of heavy metals in 0~10 cm soil layer of grassland are mostly higher than that in the remaining two layers; some of the heavy metals, such as Cu and Pb, show the pattern of decreasing and then increasing in the soil of grassland in the range of 0~10 cm, 10~30 cm, and 30~50 cm; and the contents of the heavy metals obviously change in the different sampling depths. By comparing and analyzing with the background value of Xinjiang soil [31], it can be seen that the natural grassland is most seriously polluted, and the degree of pollution of seven heavy metals can be ranked according to size as follows: Zn > Pb > Ni > Cd > Cu > Cr > As. The distribution of the heavy metals in the measured soil is consistent with the distribution of the elements in the background of soil geochemistry in Xinjiang.

Table 2. Heavy metal contents of different grassland soils in Altay.

Sampling Depth (cm)	Heavy Metal	Mean (mg·kg^{-1})					Xinjiang Background Value (mg·kg^{-1})
		No-Grazing Grassland	Seasonal Grassland	Abandoned Grassland	Natural Grassland	Artificial Grassland	
0~10	Cu	30.42	31.83	26.31	65.41	32.10	26.70
	Pb	19.58	22.38	17.87	100.08	18.98	19.40
	Cr	47.08	55.86	43.23	64.29	51.90	49.30
	Zn	71.79	75.00	68.43	408.89	72.88	68.80
	As	8.39	7.17	9.79	13.59	13.64	11.20
	Ni	52.81	36.02	60.26	106.76	52.99	25.20
	Cd	–	0.20	0.23	0.33	0.21	0.12
10~30	Cu	26.17	16.74	34.24	68.52	33.09	26.70
	Pb	16.64	16.92	17.09	89.61	18.02	19.40
	Cr	46.15	50.23	54.01	76.51	38.19	49.30
	Zn	60.28	63.58	65.27	377.04	66.09	68.80
	As	12.65	7.45	10.98	15.80	16.53	11.20
	Ni	47.70	–	47.91	66.87	46.20	25.20
	Cd	0.17	0.29	0.19	0.21	0.21	0.12
30~50	Cu	27.24	25.96	30.83	82.59	25.79	26.70
	Pb	17.76	18.73	19.08	100.13	15.03	19.40
	Cr	41.71	43.72	47.14	87.11	21.43	49.30
	Zn	60.44	63.03	64.89	470.58	57.50	68.80
	As	10.13	7.97	9.48	–	17.08	11.20
	Ni	42.81	46.58	45.38	65.55	48.65	25.20
	Cd	0.26	0.21	–	0.20	0.20	0.12

Note: "–" means not detected.

3.2. Evaluation of Soil Heavy Metal Pollution

3.2.1. Nemerow Pollution Index Method

The evaluation results of the Nemerow pollution index (Table 1) show that the mean values of P_N for the comprehensive pollution index of heavy metals in the soil of five types of grassland were 1.42 (no-grazing grassland: $1 < P_N \leq 2$, light pollution), 1.45 (seasonal grassland: $1 < P_N \leq 2$, light pollution), 1.94 (abandoned grassland: $1 < P_N \leq 2$, light pollution), 3.67 (natural grassland: $P_N > 3$, heavy pollution) and 1.63 (artificial grassland: $1 < P_N \leq 2$, light pollution). The results of the single-factor pollution index evaluation of each heavy metal element are shown in Figure 2, which indicates that the pollution of forbidden grassland is light, and the pollution of natural grassland is the most serious. As shown in (Figure 2d), the natural grassland Zn site showed 100% heavy pollution, and Cu, Pb, Ni and Cd also showed different degrees of heavy pollution.

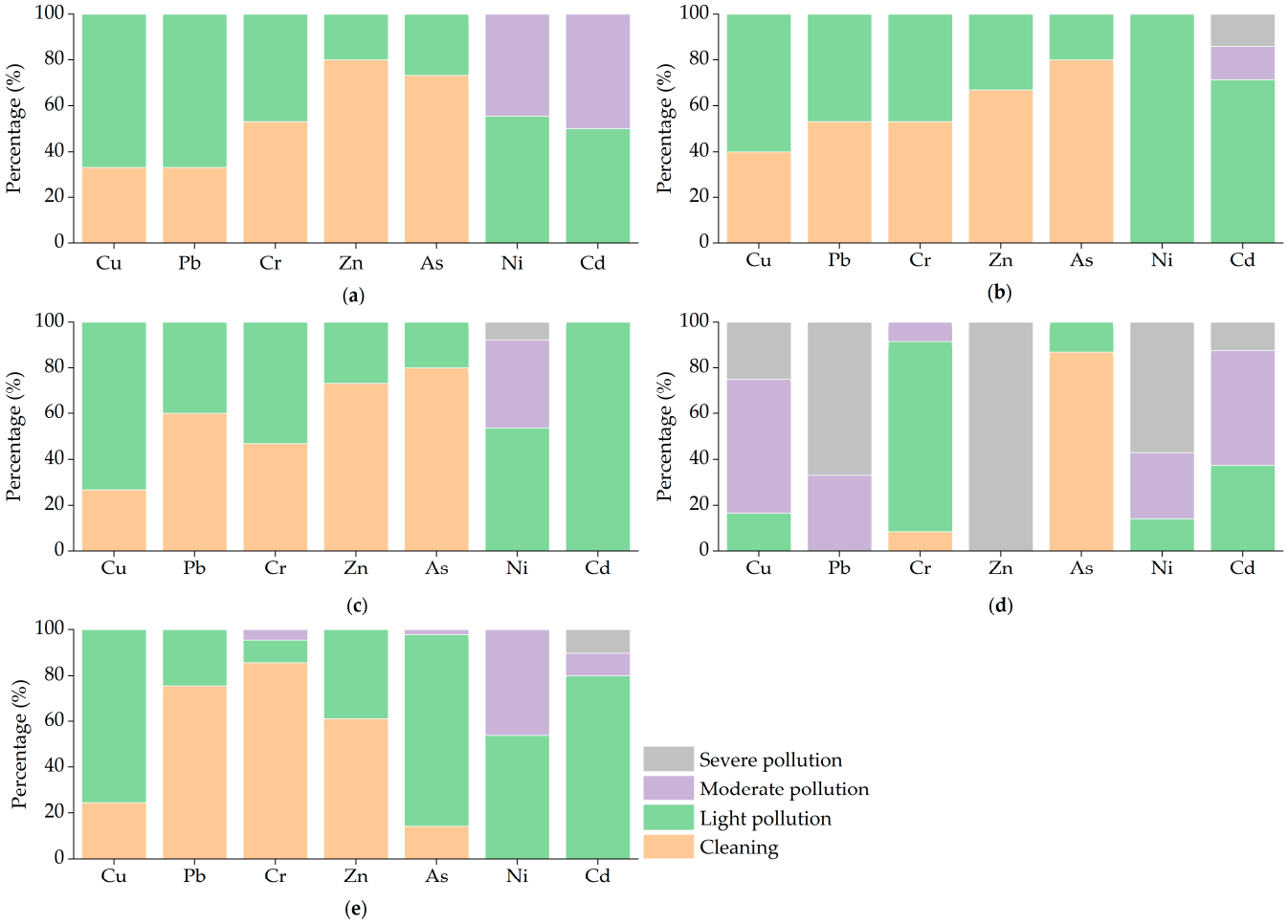


Figure 2. Evaluation results of the single-factor pollution index of different grassland soils. (**a**) Heavy metals in soil of no-grazing grassland; (**b**) heavy metals in seasonal grassland soils; (**c**) heavy metals in abandoned grassland soils; (**d**) heavy metals in natural grassland soils; (**e**) heavy metals in artificial grassland soils.

In summary, the soil heavy metal pollution in the study area was the most serious for Cd, Ni and Zn, and the regional pollution degree was as follows: natural grassland > artificial grassland > abandoned grassland > forbidden grassland > seasonal grassland. Hence, the pollution prevention and control of the above heavy metals should be strengthened.

3.2.2. Geological Accumulation Index Method

Analysis of soil contamination with grassland with metals based on I_{geo} showed that the largest contamination concerns Ni and Cd in the case of the magnitude of I_{geo} values for different heavy metals; I_{geo} values for Ni and Cd were slightly higher than 0 (Table 1 and Figure 3), which suggests that the soil is not contaminated. I_{geo} values for no-grazing grassland were slightly different. Soils were the least contaminated in no-grazing grassland, where I_{geo} values did not exceed level 1 in 100% of the samples. The most contaminated area was natural grassland, with 57% and more than 12% of samples exceeding Level 1 for Ni and Cd, respectively.

The single-factor evaluation showed that there was light Cu, Pb, Cr, Zn and As pollution at different degrees in the no-grazing grassland, seasonal grassland, and abandoned grassland, but such pollution was not according to the evaluation of the geoaccumulation index, which indicated that the levels of these five heavy metals were less affected by human activities in these types of grassland. In contrast to the single-factor index, the

results of the geoaccumulation index method did not show any heavy pollution elements, and the natural grassland showed the most obvious performance.

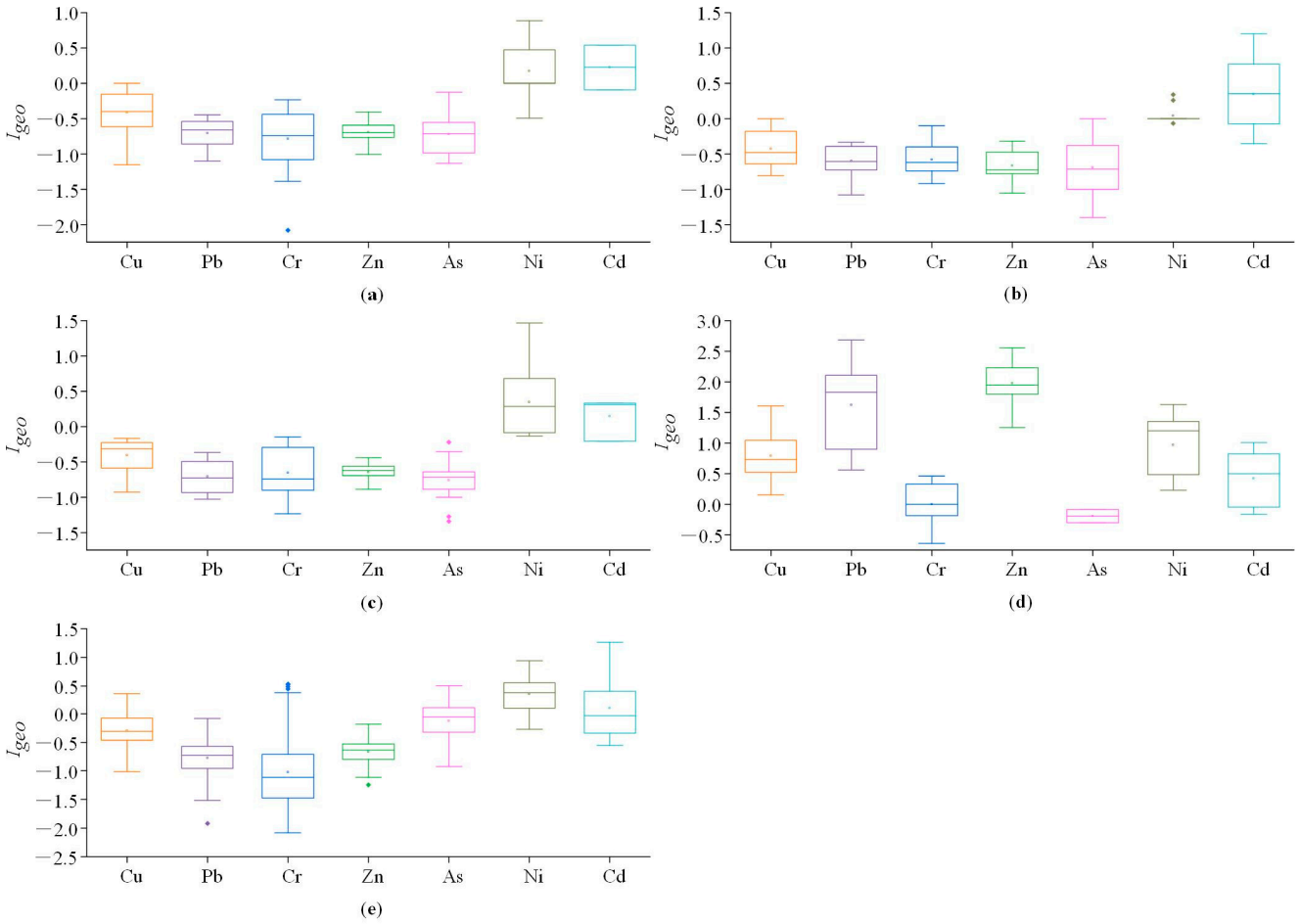


Figure 3. Evaluation results of the geological accumulation index of soil pollution in different grasslands. (**a**) Heavy metals in soil of no-grazing grassland; (**b**) heavy metals in seasonal grassland soils; (**c**) heavy metals in abandoned grassland soils; (**d**) heavy metals in natural grassland soils; (**e**) heavy metals in artificial grassland soils.

3.3. Analysis of Soil Heavy Metal Pollution Sources

3.3.1. Factor Analysis

Soil heavy metals originate from natural and human activities, and principal component analysis can effectively discern the sources of heavy metal element contamination [32]. Soil heavy metal data in the study area were standardized and subjected to KMO and Bartlett's sphere test, which showed that the KMO value was 0.767, $p = 0$ and KMO > 0.5, $p < 0.05$, indicating that the correlation of each soil heavy metal element was strong and suitable for principal component analysis; the results are shown in (Table 3). Two components had eigenvalues greater than 1, 4.143 and 1.380, respectively, with variance contributions of 59.187% and 19.709% and cumulative contributions of 78.896%. The variance of the common factor was relatively low only for Cr and As, which were 0.672 and 0.592, respectively. The others were greater than 0.750, which indicates that the overall level of the two extracted factors was high and could adequately respond to all the data.

Table 3. Results of the principal component analysis and rotational factor loadings of the soil heavy metal content.

Master Score	Eigenvalue	Contribution Rate	Cumulative Contribution Rate	Element	Factor 1	Factor 2	Common Factor Variance
F1	4.143	59.187	59.187	Cu	0.913	0.178	0.865
F2	1.380	19.709	78.896	Pb	0.906	−0.067	0.825
F3	0.533	7.612	86.508	Cr	0.806	0.151	0.672
F4	0.451	6.439	92.947	Zn	0.949	−0.008	0.900
F5	0.296	4.235	97.181	As	−0.744	0.196	0.592
F6	0.142	2.034	99.215	Ni	0.325	0.874	0.870
F7	0.055	0.785	100.000	Cd	0.529	−0.720	0.798

3.3.2. Analysis of Heavy Metal Source Contribution Based on the APCS-MLR Model for Grasslands

On the basis of the absolute scores of each principal component obtained from factor analysis, the absolute principal factor scores were analyzed via multivariate linear regression using the contents of each heavy metal, and the results showed that the R^2 values of the fit for Cu, Pb, Cr, Zn, As, Ni and Cd were 0.78, 0.82, 0.69, 0.88, 0.61, 0.90 and 0.80, respectively. The R^2 of the fit for the seven kinds of heavy metals was 0.60 or higher. The equation fit R^2 indicates the proportion of the total variation of the dependent variable in the regression relationship that can be explained via the automatic variable, and a higher R^2 means that the model is better. The R^2 results indicate that the model fit well and was suitable to be used for the study of heavy metals in Altay grassland soil.

The sum of the contribution of each source for a single heavy metal variable in the APCS-MLR model was 100%. As seen in Figure 4, the sum of the contributions of the two principal components of the seven heavy metals is less than 100%, indicating the existence of unknown sources, and such sources were related to the soil heavy metal data (selection of sampling points, number of samples, etc.) and the selected model (the model's own parameters) [33]. Combined with the factor analysis, the contributions of Cu, Pb, Cr, and Zn in source 1 were 29.7%, 61.1%, 20.2%, and 66.0%, respectively. The five types of grassland soil Cd heavy metals were more influenced by source 3, with a contribution of 67.5%, and Ni in source 2, with a contribution of 70.8%. The heavy metal As was most influenced by source 2, with a contribution of 76.7%. In addition, Pb, Zn, As and Ni were all influenced by source 3 in this study, with percentages of 38.9%, 29.5%, 23.3% and 10.2%, respectively, and their pollution sources need to be further studied, thus also revealing the diversity and complexity of the sources of heavy metal pollution in the soil of Altay grasslands.

In summary, there were three main sources of soil heavy metal pollution in the study area, as shown in (Figure 5), and the proportions were 32.6%, 24.7% and 42.7%, respectively. Combining the contribution of the three sources and the above source analysis, this shows that Pb and Zn in the study area were mainly influenced by source 1, Ni and As were mainly influenced by source 2, and Cu, Cr and Cd were mainly influenced by unknown sources.

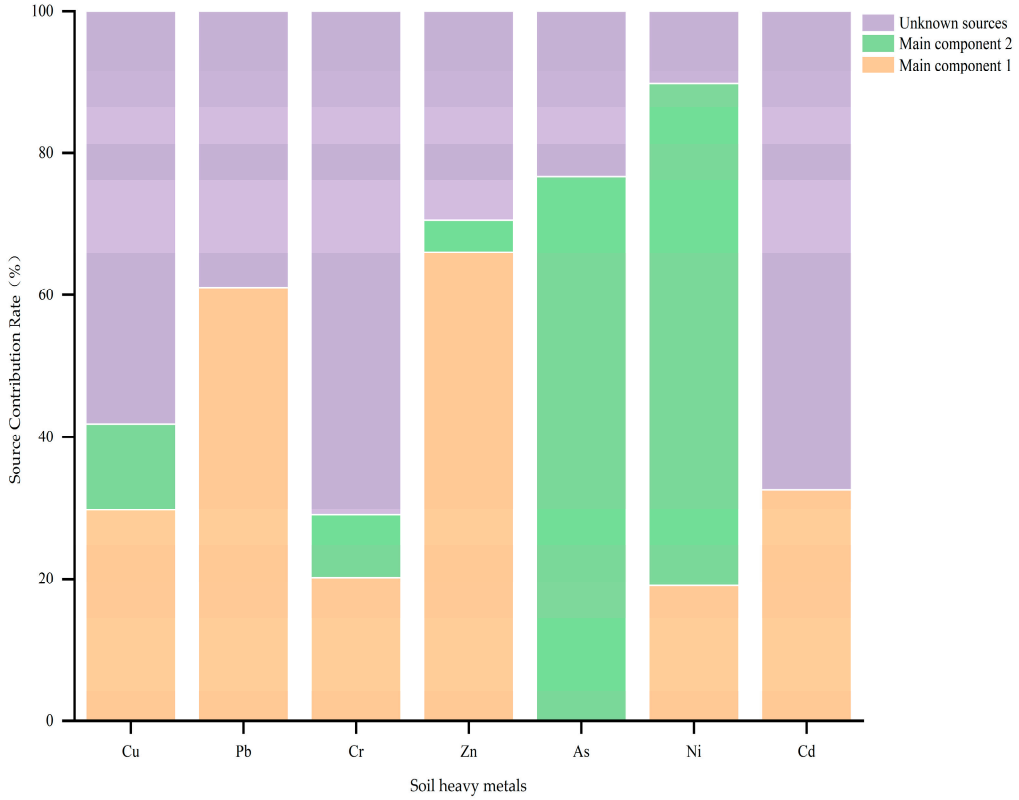


Figure 4. Contribution of heavy metal pollution sources in grassland as per the APCS-MLR model.

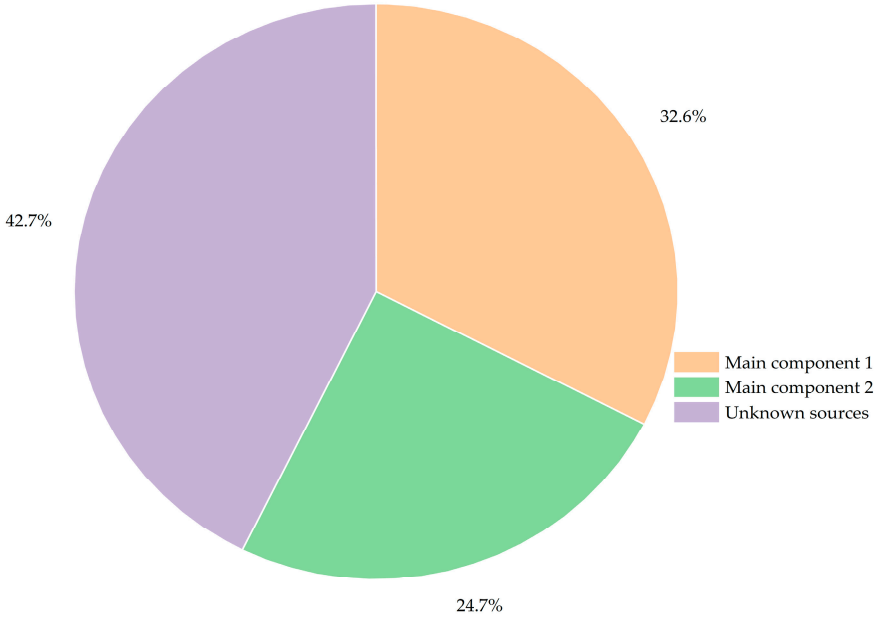


Figure 5. Average contribution of heavy metal pollution sources in grassland as per the APCS-MLR model.

4. Discussion

The contribution of principal component 1 was 59.187%, in which Cu, Pb, Cr and Zn had the largest loadings with weighting coefficients of 0.913, 0.906, 0.806 and 0.949, respectively, indicating that these four heavy metals had similar sources. Combining the mean content of heavy metals with the results of the geoaccumulation index showed that all four heavy metals in natural grasslands produced different degrees of pollution.

The Cu in the soils of closed grassland, abandoned grassland, and artificial grassland slightly exceeded the background value, and only Cr in the soils of seasonal grassland exceeded the background value. The geological accumulation index showed that artificial grassland had been lightly contaminated by Cu and Cr, and the coefficient of variation of Cr reached 65.7%, indicating that anthropogenic factors had a greater influence on Cr. According to related studies, the Altay region is mainly dominated by sandstone and volcanic rocks [34]. Yang et al. concluded that Cu and Cr in volcanic rocks mainly originate from soil parent material [35]. Wang et al. also showed that Cu and Cr mainly originate from soil parent material [36] and that Pb is mainly influenced by natural factors such as soil parent material [37]. Mineral resources are abundant in the Altay region, and mining activities not only cause serious damage to vegetation but also cause changes in the content of heavy metals in soil [34]. Du et al. studied the contamination status of soils within the Altay region, and the results showed Cu, Pb, Cr, and Zn contamination [38]. In addition, Ma et al. studied the sources of heavy metal contamination in soils around Pb-Zn mining areas, and the main source of Zn was mining activities [39]. Natural grasslands were the closest to mineral transport routes, and studies have shown that mining activities lead to the accumulation of Zn in surrounding soils [40], and in abandoned grasslands, fertilizers, agrochemical use and animal manure were a source of soil Zn [41]. The results of the one-way evaluation showed that all grassland Zn was associated with other metals such as Pb, and the risk to humans through the food chain was high. Many years of vehicle emissions [42] and mining activities [43] can lead to the accumulation of Pb in natural grasslands. Following the comprehensive analysis, principal component 1 indicates natural sources and mining activity sources as the causes of contamination.

In principal component 2, Ni had a large loading value, whereby the cumulative variance contribution was 78.896%, and its weight coefficient was 0.874. From the mean content of heavy metals and the pollution status, the mean value of soil heavy metal Ni in all five types of grassland exceeded the background values, and the evaluation results of the geological accumulation index were concentrated in clean, lightly polluted and moderately polluted soils, with relatively large variation coefficients. The maximum value of abandoned grassland was 50.3, which indicates that it was influenced by human factors. Fuyun County has a continental cold–temperate arid climate with severe and long winters, and coal-fired heating is the main heating method; there, Ni has been found to be a tracer of fuel combustion [44,45]. The industrial growth in Fuyun County has led to economic development, while at the same time bringing about environmental impacts. Thus, industrial effluent discharge has been studied as a possible source of Ni [46], and following comprehensive analysis, principal component 2 indicates industrial coal combustion and sewage sources as responsible for contamination.

Cd is strongly influenced by unknown sources, with a contribution of 67.5%. The results of the single-factor pollution and geoaccumulation index evaluations showed that Cd was more seriously polluted in natural grassland soils, which is consistent with the current types of soil heavy metal pollution in China. The study showed that Cd is a more serious metal element in soil pollution in China, specifically in the Anhui [47], Yunnan [48], Jiangxi [49], Qinghai–Tibet Plateau [50], and central [51] regions. The sources of Cd were more extensive, with agricultural fertilization [52], transportation [53], industrial activities [54], and mining [55] all contributing to different levels of Cd pollution. In this study, natural grasslands and artificial grasslands were close to the mine and transport routes, and abandoned grasslands were located near villages, which provided conditions for Cd enrichment in soils.

The results of different evaluations showed that the general heavy metal pollution of grasslands in Fuyun County, Xinjiang, was relatively light, and the moderate and above-moderate pollution was mainly distributed in the surface layer of some natural grasslands, with the Cu, Pb, Ni and Cd contents being more seriously exceeded. Accordingly, for the management of heavy metal pollution in grassland in Altay, Xinjiang, it is necessary to

strengthen the supervision of exogenous input, especially by controlling the exogenous input of excessive Zn.

5. Conclusions

(1) The heavy metal content of 0~10 cm soil in the mining area was generally higher than the remaining two layers. Natural grassland was the most seriously contaminated, and the order of its contamination was Zn > Pb > Ni > Cd > Cu > Cr > As.

(2) The comprehensive pollution index of the study area indicated that Ni and Cd were the main pollutants of the five grasslands; the accumulation of heavy metals in natural grassland soils was large, and Cu, Pb, Zn, Ni and Cd had different percentages of moderate pollution.

(3) The APCS-MLR model identified three pollution sources, and the average contributions of natural and mining activity sources, industrial coal combustion and sewage sources, and unknown sources were 32.6%, 24.7% and 42.7%, respectively.

In summary, the main pollutants in this study area are Ni and Cd, and the natural grassland is the most seriously polluted. For the remediation of soil pollution in the study area, heavy metal remediation experiments can be carried out by utilizing the plants planted in five kinds of grassland in this study, especially the needle fescue, ditch-leaved lambs' fescue, white-stemmed silky artemisia, and dogbane flower planted in the natural grassland to screen out plants, which have the capacity for enriching and transporting the soil heavy metals Ni and Cd and are resistant to drought.

Author Contributions: Conceptualization, Q.H.; methodology, Q.H.; software, Q.H.; validation, J.L., Y.W. and P.H.; formal analysis, Q.H.; investigation, Q.H.; resources, Q.H.; data curation, Q.H.; writing—original draft preparation, Q.H.; writing—review and editing, Q.H.; visualization, Q.H.; supervision, X.H.; project administration, X.H.; funding acquisition, X.H. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Key Research and Development Program of the Autonomous Region (2022B03030-2). and the National Natural Science Foundation of China (31760168).

Data Availability Statement: The datasets are available from the corresponding author upon reasonable request.

Conflicts of Interest: The authors declare no conflict of interest.

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