minerals

Article

Comprehensive Environmental and Health Risk Assessment of Soil Heavy Metal(loid)s Considering Uncertainties: The Case of a Typical Metal Mining Area in Daye City, China

Hao Zhou [1,2], Xuemei Yue [1,2], Yong Chen [1,2,3,*], Yanzhong Liu [1,3] and Gaoxu Gong [1,2]

1 School of Resources and Environmental Engineering, Wuhan University of Science and Technology, No. 947 Heping Avenue, Wuhan 430080, China; ares_zhouhao@wust.edu.cn (H.Z.); yuexuemei@wust.edu.cn (X.Y.); Liuyanzhong@wust.edu.cn (Y.L.); gonggaoxu1207@163.com (G.G.)
2 National Key Laboratory of Environmental Protection Mining and Metallurgy Resource Utilization and Pollution Control, Wuhan 430080, China
3 Hubei Provincial Key Laboratory of Efficient Utilization and Agglomeration of Metallurgical Mineral Resources, Wuhan 430080, China
* Correspondence: chenyong68@wust.edu.cn

Abstract: Heavy metal(loid)s (HMs) in soils near mining sites often cause serious environmental and health issues. Accurately assessing soil HM risks and identifying priority pollutants are crucial for improving risk control efficiency with limited management costs and resources. Traditional deterministic assessments may yield biased results due to the imprecision and ambiguity of environmental data and assessment processes. To compensate for the deficiencies of deterministic assessment, a comprehensive probabilistic-fuzzy model was developed based on fuzzy theory, probability methods, the soil contamination risk (SCR) index, and a human health risk (HR) assessment framework. According to this model, the soil HM risk status in a typical mining area in China was evaluated. The results indicated that Cd and Cu significantly violated the relevant environmental guidelines and were considered priority metals for environmental risk (ER). Notably, Cd's hazard predominantly manifested in a solid potential ecological risk (PER), whereas Cu's environmental impact primarily manifested as a soil contamination risk (SCR). From the perspective of HR, soil HMs already pose a considerable threat to human health, with children facing greater HRs than adults. As was identified as a priority element for HRs, with carcinogenic and non-carcinogenic risks reaching unacceptable levels. Regarding general risk (GR), Cd and Cu ranked in the first gradient and As in the second gradient. Overall, the accumulation of soil HMs—especially Cd, Cu, and As—in the study area has posed a significant threat to the ecosystem and human health. The risks of other HMs (Pb, Zn, Cr, and Ni) are relatively low, but the superimposed risks of multiple HMs should not be ignored. The probabilistic–fuzzy model reduces the uncertainty of risk assessment, and the model integrates the environmental and health risks of HMs, providing more comprehensive risk information. The assessment results can serve as a reference for managers to develop targeted control strategies.

Keywords: soil heavy metal(loid)s; environmental and health risks; probabilistic fuzzy model; uncertainties of risk; mining areas

check for updates

Citation: Zhou, H.; Yue, X.; Chen, Y.; Liu, Y.; Gong, G. Comprehensive Environmental and Health Risk Assessment of Soil Heavy Metal(loid)s Considering Uncertainties: The Case of a Typical Metal Mining Area in Daye City, China. *Minerals* **2023**, *13*, 1389. https://doi.org/10.3390/min13111389

Academic Editors: Pierfranco Lattanzi, Wendel Valter Da Silveira Pereira, Paula Godinho Ribeiro, Yan Nunes Dias and Antonio Fernandes

Received: 13 September 2023
Revised: 25 October 2023
Accepted: 28 October 2023
Published: 30 October 2023

Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https://creativecommons.org/licenses/by/4.0/).

1. Introduction

Soil heavy metal(loid) pollution (HMP) has emerged as a global problem in recent decades, drawing the attention of environmental managers and researchers worldwide [1–3]. Heavy metal(loid) (HM) pollutants are non-degradable, persistent, and bioaccumulative, posing a severe threat to the ecological environment and human health [4,5]. For example, the excessive accumulation of HMs can impede plant growth and decrease land productivity [6]. Exposure to HMs through ingestion, inhalation, and dermal contact can result in both non-carcinogenic and carcinogenic risks due to the potential damage that HMs can cause to humans' internal organs and nervous systems [7]. Therefore, controlling the risk of soil HMs is essential for human health and sustainable socioeconomic development.

Accurate pollution assessments can help managers determine the priority pollutants to control, thereby improving the efficiency of risk management within resource constraints [8,9]. Various methods have been developed to assess the environmental and health risks of soil HMs, such as the Nemerow integrated pollution index ($NIPI$), the potential ecological risk index (RI), and the human health risk assessment model (HHRAM) [10–12]. These methods play an essential role in the evaluation of soil HM risks. However, most previous studies have primarily employed deterministic parameters to evaluate risks when using these methods [13,14]. Uncertainty has traditionally been one of the main problems in risk assessment due to the ambiguity and imprecision of environmental data [15,16]. Ignoring uncertainties may cause one to underestimate or overestimate the actual risk [17,18]. Many researchers have gradually recognized that previous deterministic assessments may lead to biased risk management decisions [15,19,20].

Generally, uncertainty in risk assessment can be divided into two categories: stochastic and fuzzy uncertainty [21,22]. Stochastic uncertainty arises from the inherent randomness in nature, which mainly manifests in the spatial variability of HM concentrations and the exposure differences of various receptors [15,23]. Fuzzy uncertainty stems mainly from the vagueness and ambiguity in human thoughts about the parameters and results of HM risk assessment [11]. In recent years, resolving the uncertainties in risk assessment has become one of the research hotspots in soil HMs [15,24,25].

Probabilistic methods are commonly used to address stochastic uncertainty [26]. Fuzzy methods can overcome fuzzy uncertainty by using a fuzzy membership function to characterize the fuzziness of the evaluators' thoughts [27,28]. Some studies on soil HM contamination have verified the effectiveness of probabilistic and fuzzy methods in resolving uncertainties [20,29]. Stochastic uncertainty and fuzzy uncertainty often coexist in HM risk assessment [21,30]. It is difficult for a single probabilistic or fuzzy approach to simultaneously address the joint effects of stochastic and fuzzy uncertainty.

Furthermore, most existing studies tend to separate environmental and health risks when assessing soil HM risks. However, such assessments may not help the relevant authorities accurately grasp the HM risk levels and identify critical contaminants. Incomplete risk assessment results are likely to lead to poor management decisions [31,32]. For example, over-designed remediation plans may result in wasted resources and money. Moreover, the underestimation of risks may lead to inaction or limited action against pollution that causes severe damage to the natural environment and human health. Fei et al. [33] and Li et al. [34] pointed out in their study that the comprehensive assessment of ecological and health risks can provide a more accurate reference for controlling HMP.

China is rich in mineral resources [35]. The exploitation of mineral resources has caused severe damage to the ecological environment, primarily through soil HMP [11,36]. Government departments have taken active control measures. As of 2021, the exacerbation trend of soil pollution in China was initially curbed, and the overall condition of the soil environment nationwide remained stable. However, HMs continued to be the primary pollutants affecting the quality of the soil environment [37]. The weak soil HM risk management system and the severe soil HMP in mining areas are still the fundamental contradiction in China's soil eco-environmental protection. Therefore, this study proposes an integrated probabilistic-fuzzy model to address the deficiencies mentioned above regarding the existing HM risk assessment to promote the construction and improvement of the HM risk management system. First, the model can overcome stochastic and fuzzy uncertainty in risk assessments. Furthermore, the model combines environmental and health risk assessment methods, and the determined general risk can reflect the integrated impact of HMs on the environment and human health.

One of the six major Cu processing bases (Tonglushan mine) in China was selected for a case study in this study. The research results can support quantifying regional soil HM risks and the identification of priority control factors. This study mainly includes the following: (1) assessing probabilistic environmental risks and probabilistic health risks using a Monte Carlo simulation; (2) realizing a probabilistic-fuzzy environmental and health risk assessment based on probabilistic risk assessment results and a fuzzy membership function; (3) aggregating probabilistic-fuzzy environmental and health risks into probabilistic-fuzzy general risks based on a fuzzy logic reasoning system.

2. Materials and Methodology

2.1. Research Framework

This study evaluated the general risks of HMs from environmental quality and human health perspectives. Figure 1 illustrates the probabilistic-fuzzy general risk assessment framework. The framework comprises two modules: (1) a probabilistic risk assessment, which uses an algorithm from statistical sampling theory to overcome the stochastic uncertainty in the risk assessment; (2) a probabilistic-fuzzy integrated assessment based on a fuzzy reasoning system. The latter module is designed to mitigate the fuzzy uncertainty and achieve the aggregation of environmental and health risks.

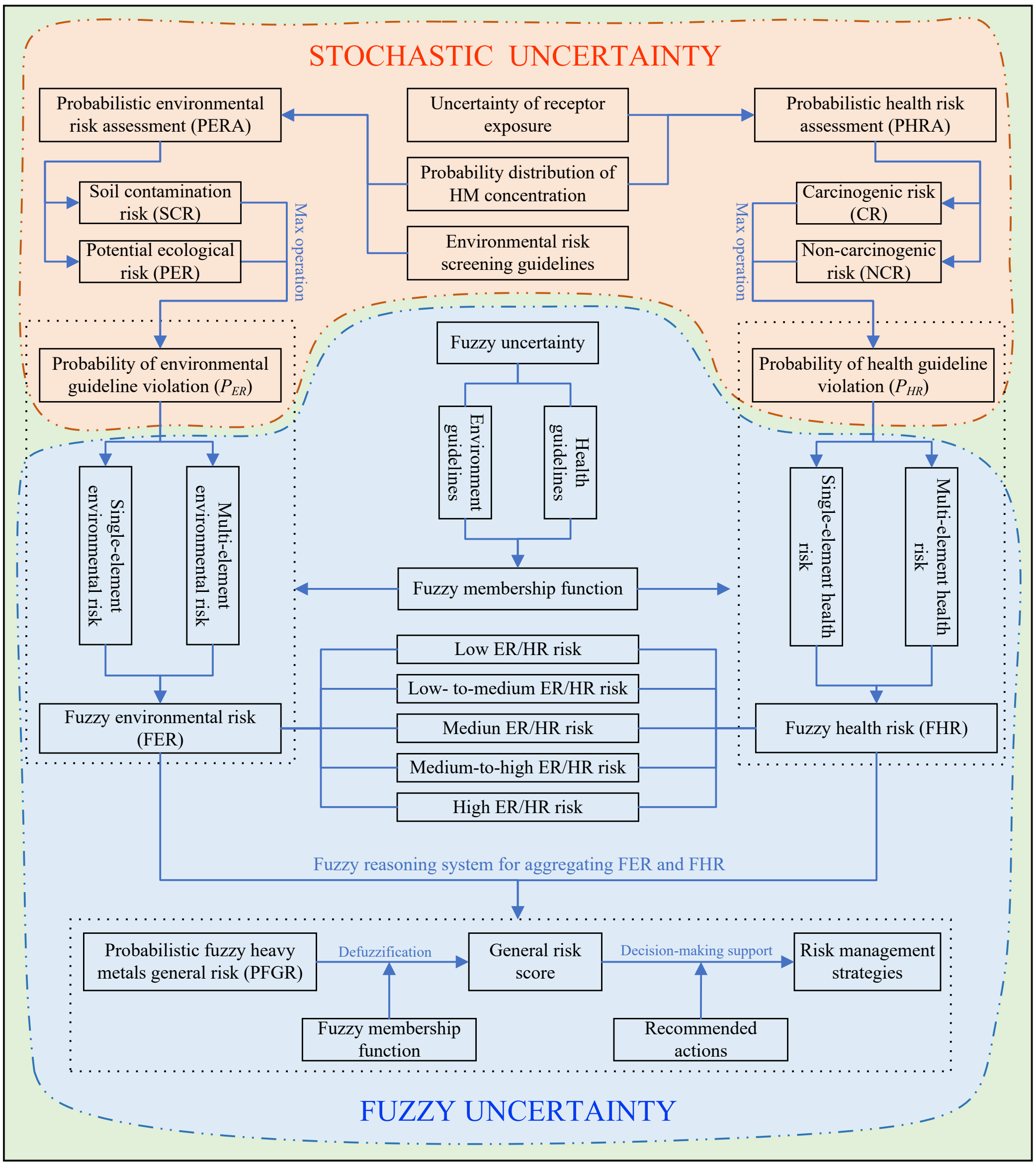

Figure 1. Theoretical framework of the probabilistic-fuzzy model.

The probabilistic risk assessment analyzes the hazards to human health and the soil environment from exposure to HMs, including a probabilistic environmental risk assessment and a probabilistic health risk assessment. Monte Carlo simulation is one of the most widely used methods for probabilistic risk assessment [38,39]. Uncertainty parameters from a Monte Carlo simulation are brought into the environmental and health risk assessment models for the probabilistic environmental and health risk assessment. The probabilities of environmental and health risks violating the corresponding guidelines set by the government are determined as the probability of environmental risk (P_{ER}) and probability of health risk (P_{HR}), respectively. P_{ER} and P_{HR} are utilized as raw data for the probabilistic-fuzzy integrated assessment.

The probabilistic-fuzzy integrated assessment consists of three parts: (1) fuzzification of the probabilistic risk assessment results; (2) aggregation of the fuzzy environmental risks (FERs) and fuzzy health risks (FHRs); and (3) defuzzification of the probabilistic-fuzzy general risks (PFGRs). First, P_{ER} and P_{HR} are fuzzified using membership functions to determine the FER and FHR. Then, the FER and FHR are aggregated into the PFGR using logic operations and the developed fuzzy rule base. Finally, the PFGR is transformed into a general risk score by the membership function (also known as "defuzzification"), thus providing more accurate and intuitive references for managers to analyze the status of HMP and determine the priority of controlling different HMs.

2.2. Probability-Based Risks

2.2.1. Environmental Risk (ER)

Soil contamination risk (SCR) and potential ecological risk (PER) are the focuses of this study on the environmental impact of soil HMs [40]. This study evaluated the environmental risks of seven mandatory HMs identified by China MEEP [41], namely Cd, As, Pb, Cr, Cu, Ni, and Zn, using the Nemerow integrated pollution index ($NIPI$) and the improved potential ecological risk index ($NIRI$) [42]. The calculations of the $NIPI$ and the $NIRI$ are shown in Equations (1)–(4). The probability of the soil contamination risk (P_{SCR}) and the probability of potential ecological risk (P_{PER}) were determined by combining the Monte Carlo simulations.

Generally, when faced with resource and budget constraints, environmental managers must prioritize mitigating factors that pose the greatest harm to environmental quality. In the environmental risk management of soil HMs, the most critical issues to be addressed are the worst effects of HMs on the soil environment. Therefore, the probability of the environmental risk (P_{ER}) is determined by the values of P_{SCR} and P_{PER}.

$$PI = \frac{C_i}{S_i} \tag{1}$$

$$NIPI = \sqrt{\frac{(PI_{i\,\mathrm{avg}})^2 + (PI_{i\,\max})^2}{2}} \tag{2}$$

$$E_r^i = T_r^i \times PI_i \tag{3}$$

$$NIRI = \sqrt{\frac{(E^i_{r\,\max})^2 + \left(E^i_{r\,\mathrm{avg}}\right)^2}{2}} \tag{4}$$

where PI and E_r^i are the single-element soil contamination and potential ecological risk indices, respectively; C_i is the HM concentration based on the probability simulation; T_r^i is the toxicity response factor of the HMs; S_i is the benchmark value for calculating PI, which is taken from the risk screening values for soil environmental quality specified by the China's MEEP [41]. $PI_{i\mathrm{avg}}$ and $PI_{i\,\max}$ are the mean and maximum values of PI for all HMs at one sampling site, respectively. $PI > 1$ indicates that the HM concentration exceeds the risk screening value specified by China MEEP. However, the risk screening value is a relatively conservative standard [41]. In previous studies [10,11], many researchers used the risk screening values developed by the MEEP as the benchmarks for calculating PI ($NIPI$) and E_r^i ($NIRI$). They pointed out that a PI ($NIPI$) value greater than 2 or an E_r^i ($NIRI$) value greater than 40 means that the threat of HMs to soil ecosystems cannot be ignored. When PI ($NIPI$) is less than 2 or E_r^i ($NIRI$) is less than 40, the risk is often considered tolerable or negligible.

Therefore, PI ($NIPI$) = 2 and E_r^i ($NIRI$) = 40 were set as the thresholds for controlling the SCR and PER, respectively. The proportion exceeding these thresholds was used to determine the probability of soil contamination risk (P_{SCR}) and the probability of potential ecological risk (P_{PER}). P_{SCR} and P_{PER} were then processed with the MAX operation to obtain P_{ER} for HMs, as shown in Equation (5):

$$P_{ER} = \mathrm{Max}(P_{SCR}, P_{PER}) \tag{5}$$

2.2.2. Health Risk (HR)

The human health risk assessment model (HHRAM) proposed by USEPA [43] was applied to evaluate the potential health risks of soil HMs. According to the model guidelines, the health risks were classified into non-carcinogenic risk (NCR) and carcinogenic risk (CR). NCR refers to the non-carcinogenic health effects of chronic exposure, such as teratogenic and genetic effects [44]. CR is the incremental probability that an individual will develop lifelong cancer due to exposure to carcinogenic HMs [45].

This study considered three exposure pathways: oral ingestion, dermal exposure, and inhalation [46]. The Monte Carlo method was used to simulate uncertain parameters in the health risk model [20,47]. The details of the HHRAM and calculation parameters are presented in Text S1 and Tables S1–S3 in the Supplementary Material.

Mining areas are sites with a high incidence of soil HMP. We determined the thresholds for controlling HRs caused by HMs based on the technical guidelines for risk assessment proposed by China MEEP [48]. The NCR was considered unacceptable when the HI (THI) exceeded 1. Meanwhile, a value of CR or TCR above the risk threshold 5×10^{-5} indicated an unacceptable carcinogenic risk. Therefore, we determined the probability of non-carcinogenic risk (P_{NCR}) and carcinogenic risk (P_{CR}) based on the probability distributions of the HI and CR (threshold: HI = 1, CR = 5×10^{-5}).

The NCRs of seven HMs and the CRs of four HMs (As, Pb, Cr, and Ni) were evaluated in this study. These four HMs have been widely recognized as causes of CR in previous studies and identified by the MEEP as intervention elements for agricultural soils [41,49]. The health risks were separately assessed for adults and children (6–18 years), considering the physiological and behavioral differences in the population. Finally, higher values of P_{NCR} and P_{CR} for adults and children were determined as the probability (P_{HR}) of violating the health guidelines, as shown in Equation (6). The P_{ER} and P_{HR} values obtained from the probabilistic environmental and health risk assessment were further processed with fuzzy techniques to quantify the probabilistic-fuzzy general risks of the HMs.

$$P_{HR} = \mathrm{MAX}(P_{NCR}, P_{CR}) \tag{6}$$

2.3. Probabilistic–Fuzzy General Risk (PFGR) Assessment

2.3.1. Fuzzification of the Probabilistic Risk

There is ambiguity in an assessor's perception of the risk assessment index, which leads to fuzzy uncertainty in the risk results. Therefore, this study used fuzzy techniques to overcome the ambiguity and imprecision of human judgment in the risk assessment process. The fuzzification of risk is the first step in resolving the uncertainty. The fuzzy membership function can systematically transform human perception or linguistic variables into the corresponding membership levels. According to the fuzzy membership function, P_{ER} and

P_{HR} are mapped to different fuzzy risk sets to determine the fuzzy environmental and health risks. A fuzzy set can be expressed as follows.

$$F(x) = \{(x, \mu_F^x), x \in X\}, \mu_F^x : X \to [0, 1] \tag{7}$$

where X is a universal set of variable x, $F(x)$ is a fuzzy set of X, and μ_F^x is the degree of membership for x in $F(x)$. The value of μ_F^x ranges from 0 to 1, and a higher value of μ_F^x indicates a stronger association between x and $F(x)$. Fuzzy sets are represented by membership functions. Figure S1 shows the triangular and trapezoidal membership functions defined by fuzzy numbers (a, m, b) and (a, m, n, b).

The fuzzy membership function is an important means of achieving the fuzzification of risk assessment results. We developed a fuzzy membership function for the probabilistic environmental and health risks by referring to relevant risk guidelines and survey reports [50,51], as shown in Figure S2. This fuzzy membership function can map P_{ER} and P_{HR} to five fuzzy risk sets: low (L), low–moderate (L-M), moderate (M), moderate–high (M-H), and high (H). The membership levels of P_{ER} and P_{HR} in the different risk sets represent the FER and FHR. For example, if the calculated probability of a multi-element integrated risk violating environmental guidelines is 0.70 ($P_{ER} = 0.7$), the environmental risk can be partially considered as M-H ($\mu_{M\text{-}H}^{ER} = 0.50$) and H ($\mu_H^{ER} = 0.50$). Therefore, the FER can be expressed as ($\mu_{M\text{-}H}^{ER} = 0.50, \mu_H^{ER} = 0.50$). If the calculated probability of violating the health guidelines for a single element is 0.85 ($P_{HR} = 0.85$), the health risk of the element can be fully determined to be H ($\mu_H^{HR} = 1.00$).

2.3.2. Fuzzy Risk Aggregation and De-Fuzzification

The purpose of constructing the indicator of "general risk (GR)" is to characterize the comprehensive risk of HMs to the environment and health. Currently, there is no quantitative mathematical model for combining these two risks. This study used fuzzy reasoning to determine the GRs of the HMs. The fuzzy reasoning process included qualitative and quantitative methods. First, a fuzzy rule base was developed based on expert judgments and related research results, and it contained 25 fuzzy rules, as shown in Table S4. As mentioned in Section 2.3.1, each value of P_{ER} and P_{HR} was mapped to one or two fuzzy risk sets so that up to four different "fuzzy environmental risk-fuzzy health risk" (FER–FHR) combinations could be generated. The developed fuzzy rules qualitatively determined the general risk level of these FER–FHR combinations. The aggregated probabilistic-fuzzy general risks (PFGRs) were identified according to five fuzzy levels: low (L), low–moderate (L-M), moderate (M), moderate–high (M-H), and high (H), as shown in Figure S3.

Fuzzy rules can only qualitatively determine the fuzzy level of the PFGR. The membership level of the PFGR in a fuzzy risk set is determined by fuzzy logic operations. The whole reasoning process consists of two fuzzy logic operations. First, the AND operation determines the membership level of the PFGR formed by different FER–FHR combinations. Then, the OR operation aggregates all PFGRs formed by the FER–FHR combinations into the final PFGR. The final PFGR is represented as a new combined fuzzy set.

The representational form of the fuzzy set cannot provide intuitive references for risk control. A de-fuzzification operation transforms the final combined fuzzy set for the PFGR into a general risk score of 0–100. As shown in Equation (8), the horizontal coordinate of the gravity-based centroid of the final fuzzy set for the integrated general risk is determined as the general risk score. The lower the score, the lower the general risk of HMs in the region. The relevant departments can directly judge the status of HMP in the soil based on the general risk score and can identify priority pollutants. This study suggests HMP control measures according to the general risk score, as shown in Table S5.

$$\text{Score}_{GR} = \frac{\int x\mu_F^x dx}{\int \mu_F^x dx} \tag{8}$$

2.4. Study Area and Materials

2.4.1. Study Area and Sampling

The mining area investigated in this study is located in Tonglushan, Daye City, Hubei Province, and is one of the world's oldest mining regions. The mining area hosts large-scale and high-grade mineral resources, primarily copper-rich and iron-rich ores, such as copper ore, copper–iron ore, iron ore, and copper–sulfur ore. The ore minerals include chalcopyrite, pyrite, bornite, chalcocite, magnetite, and hematite. The stratigraphy within the study area predominantly originated from the Early Cretaceous and Holocene. The central region of the study area is dominated by Early Cretaceous quartz diorite, while the periphery consists of Holocene lacustrine deposits and alluvial sediments. Mining activities in the Tonglushan mine can be traced back to approximately 3000 years ago. The mining industry's development has propelled the region's socio-economic growth; however, it has unavoidably caused severe soil HMP. Through preliminary field surveys, 17 villages within 3 km of Tonglushan were delineated as within the formal investigation area. Soil sampling was conducted in December 2020.

The soil sampling process strictly adhered to the "Technical Specification for Soil Environment Monitoring (HJT 166–2004)" developed by China MEEP [52]. Before sampling, we prepared the sampling work maps and the necessary tools for soil collection. This sampling primarily focused on agricultural soils, and the region mainly cultivated common crops, so we collected soil samples from the tillage layer (0–20 cm). Given the hilly terrain prevalent in the study area, each sampling point was set up with 10–15 subpoints using the S-shaped layout. Soil from each subpoint was thoroughly mixed, and a 1 kg composite sample was obtained using a quartering technique. The collected composite soil samples were placed into polyethylene sample bags, labeled, and registered in a sample log. A handheld GPS device (Garmin GPSMAP 631sc, Switzerland) was employed to record the geographical coordinates (as shown in Figure S4) and sampling timestamps for each sample. Sixty composite soil samples were gathered and stored in soil storage containers and transported to the laboratory.

2.4.2. Chemical Analysis

After air-drying the soil samples in the laboratory, the removal of stones and debris was carried out. Subsequently, the dried soil samples were ground and passed through a 20-mesh sieve to obtain the test soil samples. Following the guidelines specified by China's MEEP, the pH of all samples was determined using a pH meter (Leici, PHSJ–3F, Shanghai, China). After completing the pH measurements, the soil samples underwent complete digestion using aqua regia (3:1 HCl/HNO_3). The digestion process was conducted in a microwave digestion apparatus (Sineo, SH60A, Shanghai, China) and consisted of three steps. First, the temperature was raised to 120 °C within 7 min and held for 3 min. Then, the temperature was raised to 160 °C within 5 min and held for 3 min. Finally, the temperature was raised to 190 °C within 5 min and held for 25 min. An electric hotplate (Sineo, ECH–20, Shanghai, China) was employed to drive the acidity of the digested liquid to obtain the test solution. Subsequently, heavy metal(loid) concentrations were measured using inductively coupled plasma mass spectrometry (ICP–MS, Agilent 7500C, Santa Clara, CA, USA) [53].

Regarding quality assurance/quality control (QA/QC), analysis was conducted on blank samples, duplicate samples, and standard reference materials. The standard reference materials used, GSS–11, GSS–13, GSS–14, GSS–18, and GSS–19, were sourced from the China National Research Center for Geoanalysis. The differences between the detected concentrations and certified values of the standard materials were all less than 10%. The spike recoveries fell within the range of 96.3%–106.2%. The relative deviations for duplicate samples were all less than 5%. Three measurements were retained for each sample, and the average value was taken as the final analytical result. This study primarily focused on the mandatory elements for screening agricultural land pollution risk in China (GB15618–2018) [41], which include Cd, As, Pb, Cr, Cu, Ni, and Zn. Following the analysis of all composite soil samples, one outlier sample was removed, resulting in the retention of 59 analytical results, as shown in Supplementary Material Table S6.

3. Results and Discussion

3.1. Probabilistic Environmental Risk Assessment

This study analyzed the environmental risk from two perspectives: soil contamination risk (SCR) and potential ecological risk (PER). The assessment results can reflect the hazards of HMs to soil ecology and the extent to which HMs violate environmental guidelines. The probabilistic environmental risk assessment of the HMs was completed by combining the Monte Carlo model and the environmental risk indexes. As mentioned in Section 2.2.1, $PI(NIPI) = 2$ and $E_r^i(NIRI) = 40$ were the thresholds for controlling the SCR and PER, respectively. To facilitate the comparative risk analysis, we mapped the critical values of the SCR and PER to 1.0, and the remaining values were mapped in equal proportions.

Table 1 summarizes the descriptive statistics of the probabilistic environmental risk assessment results. Regarding the single-element risk, Cu and Cd were the HMs with the highest SCR and PER values, respectively. Their mean and median values exceeded the corresponding thresholds of risk control. The probability of soil contamination risk (P_{SCR}) for Cu was 62.34%. The probability of potential ecological risk (P_{PER}) for Cd was 64.55%. One of China's six productive Cu mines (Tonglushan mine) is in this study area. With the support of abundant mineral resources, a complete mining–beneficiation–smelting system was established.

In our previous traceability research conducted on soil HMs in this region, Cu and Cd were identified as indicative elements of mining and smelting industrial sources, respectively, [40]. Copper ore is the most dominant product of the mine. Frequent mining operations lead to a significant migration of HMs—especially Cu—into the soil environment [54]. The combustion of fossil fuels often exacerbates the accumulation of Cd in the soil. Cd is typically present in trace amounts in fossil fuels, especially coal [55], and smelting operations in the study area require a substantial amount of coal as an energy source. Cd from coal can adhere to coal ash and waste particles during the smelting process, consequently accumulating in the soil. In addition, Cd is also a commonly associated element in copper ores [56]. The improper disposal of waste during ore processing can lead to an increase in Cd concentration in the soil. The combined influence of these factors resulted in a significantly higher environmental risk for Cu and Cd in this region compared to other HMs.

Table 1. Descriptive statistics of the probabilistic environmental risk assessment results.

Element	Soil Contamination Risk (SCR)			P_{SCR} (%)	Potential Ecological Risk (PER)			P_{PER} (%)	P_{ER} (%)
	Mean	Median	90th P		Mean	Median	90th P		
As	0.77	0.66	1.33	23.12	0.39	0.33	0.67	2.49	23.12
Cd	1.61	0.96	3.50	48.05	2.36	1.44	5.16	64.55	64.55
Cr	0.22	0.21	0.32	0.00	0.02	0.02	0.03	0.00	0.00
Pb	0.35	0.25	0.71	4.54	0.09	0.06	0.18	0.00	4.54
Cu	1.91	1.32	3.95	62.34	0.48	0.33	0.98	9.56	62.34
Ni	0.13	0.11	0.23	0.00	0.03	0.03	0.06	0.00	0.00
Zn	0.46	0.39	0.81	4.92	0.02	0.02	0.04	0.00	4.92
Total	2.01	1.47	3.81	73.60	1.76	1.10	3.71	54.41	73.60

Except for Cu and Cd, only the 90th percentile of the SCR for As exceeded the risk control threshold. Compared to these elements (Cu, Cd, and As), the other HMs were at relatively low environmental risk. From the multi-element total risk perspective, the mean and median values of SCR and PER exceeded the corresponding risk thresholds, and the total SCR was higher than the total PER. The sensitivity analysis of the Monte Carlo simulation showed that Cu and Cd contributed 62% and 37% of the variance in the multi-element total SCR assessment, respectively. Moreover, Cd contributed 98% of the variance in the multi-element total PER. In summary, Cu and Cd were the primary elements to be controlled in the study area regarding environmental risk.

Figure 2a–g show the probability distribution of the single-element SCR and PER. It can be seen that the SCR of Cr and Ni and the PER of Cr, Pb, Ni, and Zn did not exceed 1 (after equal proportional mapping). Figure 2h shows the probability distributions of the total SCR and PER for the seven elements. The probability distribution curve can intuitively reflect the probability of the SCR and PER exceeding the risk control threshold. P_{SCR} and the P_{PER} were substituted into Equation (5) to determine the probability of environmental risk (P_{ER}).

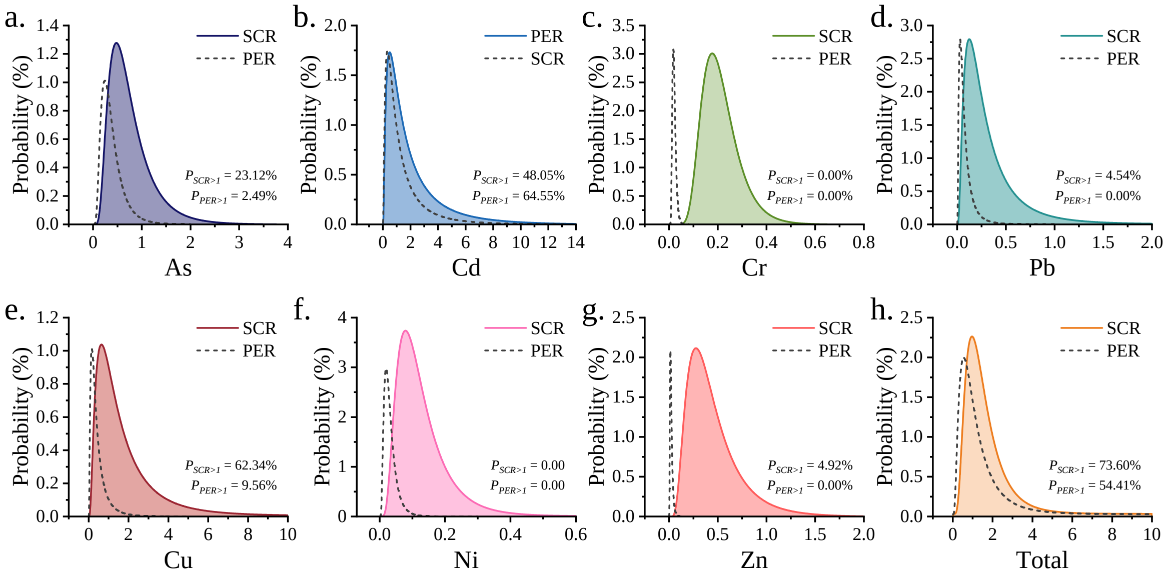

Figure 2. Single-element (**a**–**g**) and multi-element (**h**) probabilistic soil contamination risks and potential ecological risks.

As shown in Table 1, the environmental risk of Cr and Ni was negligible. The P_{ER} values of the five remaining elements decrease in the following order: Cd (64.55%) > Cu (62.34%) > As (23.12%) > Zn (4.92%) > Pb (4.54%). The probability of multi-element total environmental risk (P_{TER}) for the seven elements was 73.60%. In Section 3.3, the single-element P_{ER} and multi-element P_{TER} are mapped to different fuzzy risk sets according to the fuzzy membership function in Figure S2 and, thus, transformed into probabilistic-fuzzy environmental risks.

3.2. *Probabilistic Health Risk Assessment*

Table 2 summarizes the probabilistic health risk assessment results for non-carcinogenic and carcinogenic effects. It can be seen that children face higher NCR and CR than adults. Comparing the NCR of the seven HMs, we found that the mean and median HI values for adults and children decreased in the following order: As > Cr > Pb > Cu > Cd > Ni > Zn. The mean and median CR values for four carcinogenic elements were ranked as follows: As > Cr > Cd > Pb. Compared with their NCR rankings, the CRs of As and Cr remained the same, but Pb and Cd swapped places. We speculate that this was related to differences in the processing of the exposure pathway for Pb in the NCR and CR assessments. The relevant guidelines do not provide the carcinogenic slope factor for Pb through the dermal pathway. Some previous studies also did not consider the CR of Pb via the dermal route. Therefore, in this study, the carcinogenic effects of Pb via the dermal route were temporarily ignored. However, the NCR assessment of Pb did consider the risk via the dermal route, which may be the main reason for the lower ranking of Pb in the CR assessment. However, it was undisputed that As posed the highest health risk.

Table 2. Descriptive statistics of non-carcinogenic and carcinogenic health risks based on the Monte Carlo simulation.

HMs	Age	Non-Cancer Risk (NCR, Represented by HI)			Cancer Risk (CR)		
		Mean	Median	90th Percentile	Mean	Median	90th Percentile
As	Children	5.97×10^{-1}	4.12×10^{-1}	1.08×10^{0}	2.85×10^{-5}	1.77×10^{-5}	5.94×10^{-5}
	Adults	1.24×10^{-1}	1.02×10^{-1}	2.24×10^{-1}	2.06×10^{-5}	1.52×10^{-5}	4.45×10^{-5}
Cd	Children	7.56×10^{-3}	3.19×10^{-3}	1.49×10^{-2}	3.30×10^{-6}	1.19×10^{-6}	6.58×10^{-6}
	Adults	1.59×10^{-3}	8.61×10^{-4}	3.60×10^{-3}	1.39×10^{-6}	6.00×10^{-7}	3.26×10^{-6}
Cr	Children	1.16×10^{-1}	8.78×10^{-2}	1.95×10^{-1}	1.85×10^{-5}	1.25×10^{-5}	3.49×10^{-5}
	Adults	2.26×10^{-2}	2.03×10^{-2}	3.60×10^{-2}	1.11×10^{-5}	9.17×10^{-6}	2.24×10^{-5}
Pb	Children	9.03×10^{-2}	5.07×10^{-2}	1.79×10^{-1}	2.35×10^{-7}	1.35×10^{-7}	6.11×10^{-7}
	Adults	1.23×10^{-2}	8.12×10^{-3}	2.59×10^{-2}	1.21×10^{-7}	6.78×10^{-8}	2.81×10^{-7}
Cu	Children	2.77×10^{-2}	1.52×10^{-2}	5.92×10^{-2}			
	Adults	3.64×10^{-3}	2.26×10^{-3}	7.81×10^{-3}			
Ni	Children	5.22×10^{-3}	3.82×10^{-3}	9.11×10^{-3}			
	Adults	7.55×10^{-4}	6.52×10^{-4}	1.33×10^{-3}			
Zn	Children	2.85×10^{-3}	1.77×10^{-3}	5.12×10^{-3}			
	Adults	3.66×10^{-4}	2.77×10^{-4}	7.19×10^{-4}			
Total	Children	8.47×10^{-1}	6.10×10^{-1}	1.45×10^{0}	5.06×10^{-5}	3.40×10^{-5}	9.78×10^{-5}
	Adults	1.66×10^{-1}	1.43×10^{-1}	2.80×10^{-1}	3.32×10^{-5}	2.69×10^{-5}	6.77×10^{-5}

Overall, NCR and CR's mean and median values for all elements do not exceed the respective risk thresholds (HI: 1.0×10^{0}, CR: 5.0×10^{-5}). Only the 90th percentile of As risk exposure for children reached 1.08×10^{0} (HI) and 5.94×10^{-5} (CR). Figure 3a,b show the cumulative probability distribution curves of the single-element NCR and CR, respectively. The probability distribution characteristics indicated that all single-element NCRs faced by adults were acceptable.

The statistics showed that about 11.69% of the As NCRs for children exceeded the guideline threshold. The NCRs of all other HMs for children and adults were below the corresponding thresholds. In addition, about 7.41% and 13.58% of the As CRs for adults and children, respectively, exceeded 5.0×10^{-5}, and about 4.71% of the Cr CRs for children exceeded 5.0×10^{-5}. These risks must be taken seriously and controlled.

On the one hand, tracing the sources of HMs in the soil can enable the development of targeted management strategies for controlling key risk elements, thus preventing the continued accumulation of HMs at the source. It is worth noting that controlling the sources addresses the issue of incremental HMs in the soil. Soil remediation is key to solving the existing stock of HMs in the soil. Local governments must actively engage in and promote soil management activities, such as the passivation of HMs or the introduction of specific plants to absorb HMs from the soil and their subsequent centralized treatment.

The CR for Cd was acceptable, with a 90th percentile above 1.0×10^{-6} but less than 5.0×10^{-5}. Meanwhile, the 90th percentile of the Pb CR was less than 1.0×10^{-6}, which was negligible. In summary, the single-element health risks of the other HMs were acceptable or negligible, except for As and Cr.

The multi-element integrated health risk is also an essential indicator for risk control due to the superimposed effect of heavy-metal hazards [49]. As reported by Yang et al. [57], the multi-element integrated risk may exceed the guideline thresholds even if all single-element health risks do not violate health guidelines. Table 2 shows that the 90th percentile of the multi-element total health risk exceeded the corresponding thresholds for all exposure scenarios except for the adult NCR.

Figure 3c,d illustrate the cumulative probability distribution curves for the multi-element total non-carcinogenic risk (TNCR) and the total carcinogenic risk (TCR). It can be seen that the multi-element TNCR faced by adults was acceptable (THI < 1). However, about 20.78% of the TCR for adults was unacceptable. For children, about 22.42% and 33.03% of the TNCR and TCR, respectively, exceeded the tolerable limits.

As described earlier, children were at a higher health risk than adults, similar to the findings of some previous studies on the health risk assessment of soil HMs [58]. The health risk assessment model indicates that children's lower body mass is one of the reasons why they are at higher risk. Additionally, children's lower resistance to HMs, as well as their geophagia and xenophagia, may also be associated with their higher risk [8,59]. Parents should monitor their children to avoid excessive contact with contaminated soil and improve their child's hygiene. These measures will effectively reduce their potential health risks from soil contamination.

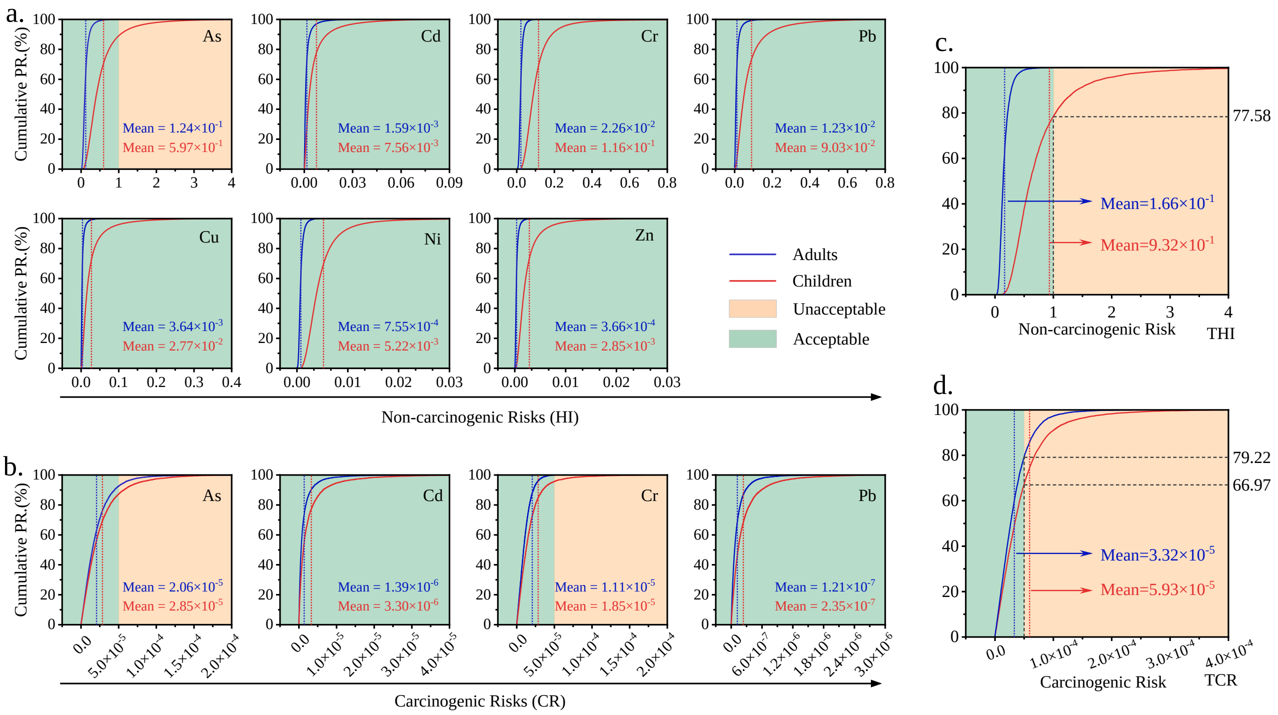

Figure 3. Cumulative probability distribution of health risks: single-element non-carcinogenic risk (**a**), single-element carcinogenic risk (**b**), multi-element total non-carcinogenic risk (**c**), multi-element total carcinogenic risk (**d**).

The NCR and CR were processed with the MAX operation to determine the probability of health risk (P_{HR}), as shown in Equation (6). The results showed that the P_{HR} values for As and Cr were 13.58% and 4.71%, respectively. The probability of the multi-element total health risk was 33.03%. Comparing the assessment results for the environmental and health risks, we observed an interesting phenomenon. Cu and Cd had the highest environmental risk but did not pose serious health risks. This is because the toxicity coefficient of Cu in humans is relatively low. Furthermore, although Cd has a higher toxicity coefficient, Cd in soil is not easily absorbed into the human body through various exposure routes such as oral ingestion, dermal exposure, and inhalation. On the contrary, As and Cr are more likely to cause harm to the human body through these three exposure pathways than Cd. In particular, As can easily enter the human body through skin contact and inhalation. This is the primary reason why As and Cr, despite their relatively lower environmental risks, result in greater health risks. As Yuan et al. [60] suggested, the HMs that contribute less to soil contamination pose the most significant health risks. This phenomenon also reflects the heterogeneity of the risks of different HMs [33]. Therefore, it is necessary to consider the comprehensive hazards of HMs when developing risk control strategies.

The adverse effects of HMs in soils are complex and widespread. Compared to urban and coal mining areas, soil HMP in non-ferrous metal mining areas is often more severe and difficult to manage [8]. Currently, scholars have provided abundant technical support for controlling HMP. However, management costs (e.g., money and time) are still significant factors limiting the control of HMP [61,62]. The lack of information on HM risks can lead to a waste of resources in the risk control process. To increase the effectiveness of unit costs, managers must fully consider the comprehensive environmental and health impacts of HMs when developing strategies [32,63]. In Section 3.3, we will assess the general risk of HMs using fuzzy methods based on the probabilistic environmental and health risk assessment results.

3.3. General Risk Based on Probabilistic–Fuzzy Assessment

The first step in the general risk (GR) assessment is to transform the probabilistic environmental and health risks into fuzzy environmental risk (FER) and fuzzy health risk (FHR). According to the fuzzy membership function in Figure S2, the P_{ER} and P_{HR} values determined in the probabilistic environmental and health risk assessment were mapped to different fuzzy risk sets. Then, the FER and FHR were aggregated into general risk through fuzzy reasoning.

Taking As as an example, the P_{ER} and P_{HR} values were determined to be 23.12% and 13.58% in the probabilistic environmental and health risk assessments, respectively. Figure S2 shows that the risks of As were mapped to the environmental risk fuzzy sets of L-M and M and the health risk fuzzy sets of L and L-M. The membership levels of As in different fuzzy sets were calculated according to the membership function in Figure S1. The fuzzy environmental risk ($\mu_{L\text{-}M}^{ER} = 0.85$, $\mu_{M}^{ER} = 0.15$) and fuzzy health risk ($\mu_{L}^{HR} = 0.30$, $\mu_{L\text{-}M}^{HR} = 0.70$) were included. Thus, four different FER–FHR combinations were generated (as shown in Figure 4).

The general risk levels of different FER–FHR combinations could be qualitatively determined from the fuzzy rule base in Table S4. The AND operation was used to calculate the membership level of the GR obtained from the FER–FHR combinations in the fuzzy set, as shown in Figure 4. For example, when environmental risks were mapped to the fuzzy set of L-M ($\mu_{L\text{-}M}^{ER} = 0.85$) and health risks were mapped to the fuzzy set of L ($\mu_{L}^{HR} = 0.30$), the general risk was determined to be L-M ($\mu_{L\text{-}M}^{GR} = 0.30$). Figure 4c,f,i,l show the general risk fuzzy sets aggregated from the four different FER–FHR combinations for As.

These four general risk fuzzy sets were then aggregated into the final combined fuzzy set of probabilistic-fuzzy general risk (PFGR) with the fuzzy OR operation, as shown in Figure 4m. The horizontal coordinate of the gravity-based centroid of the final combined fuzzy risk set (Figure 4m) was determined as the general risk score for As. According to Equation (8), the final general risk score for As was calculated to be 32.86. As suggested in Section 2.3.2, this general risk score indicated that further investigation should be conducted to determine more detailed information on As contamination.

The same fuzzy reasoning methods were used to quantify the general risk of other HMs. Figure 5 shows the final combined fuzzy sets of PFGR and the general risk scores for all HMs except for As. The multi-element total general risk score was 83.34, indicating that the agricultural land in the study area requires comprehensive control and remediation measures for HMP.

In terms of single-element risk, Cd was the element with the highest general risk. The strong biotoxicity of Cd makes it a high-potential ecological risk to the environment. The general risk of Cu was close to that of Cd. Although the biotoxicity of Cu is weaker than that of other HMs, the accumulation of Cu in the soil was significant due to the presence of a large Cu mine in the study area. Therefore, managers still need to pay attention to the risks of Cu.

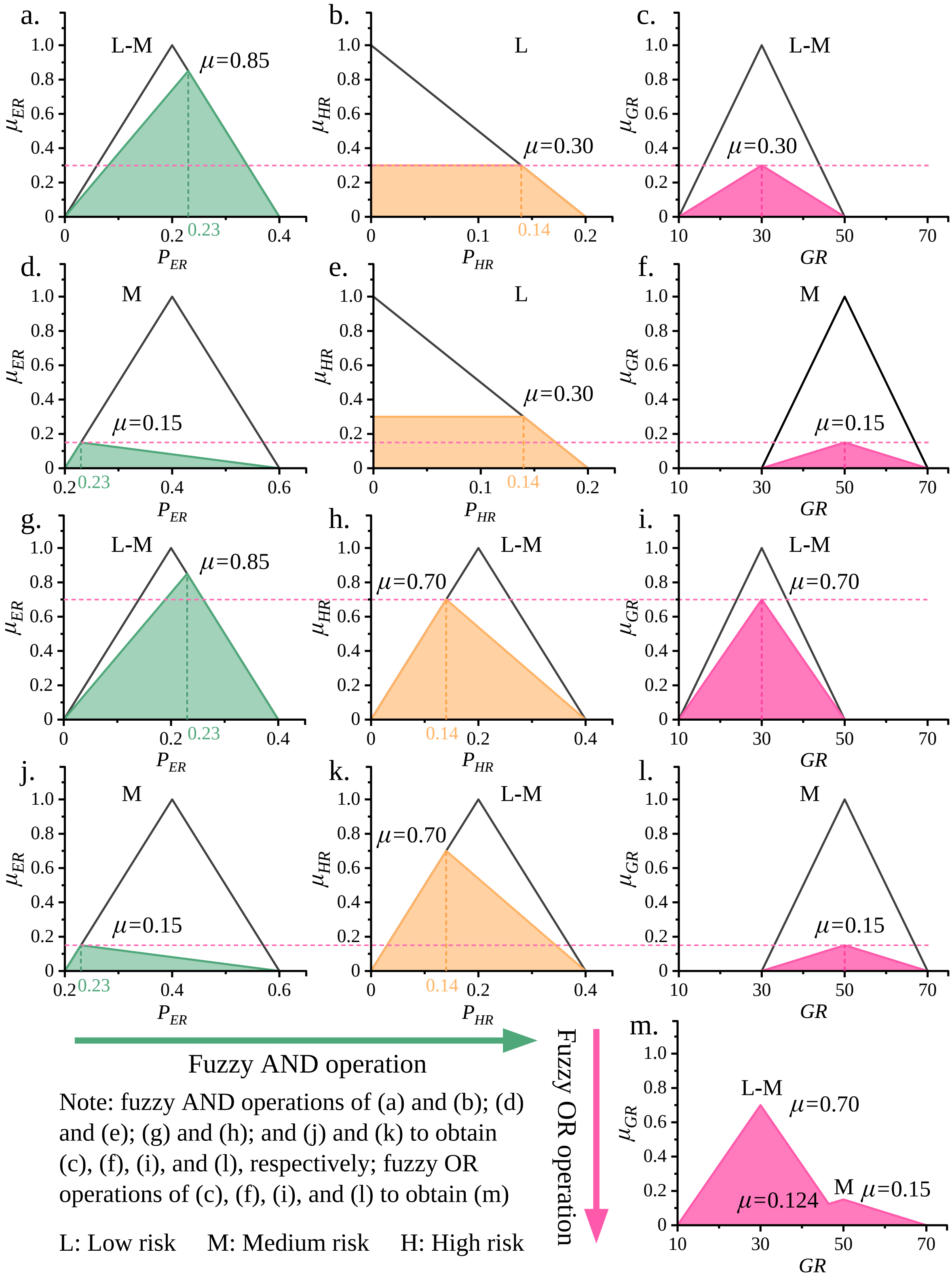

Figure 4. Probabilistic fuzzy general risk of As. Probabilistic fuzzy general risk of As. Note: fuzzy AND operations of (**a**,**b**,**d**,**e**,**g**,**h**,**j**,**k**) to obtain (**c**,**f**,**i**,**l**), respectively; fuzzy ORoperations of (**c**,**f**,**i**,**l**) to obtain (**m**).

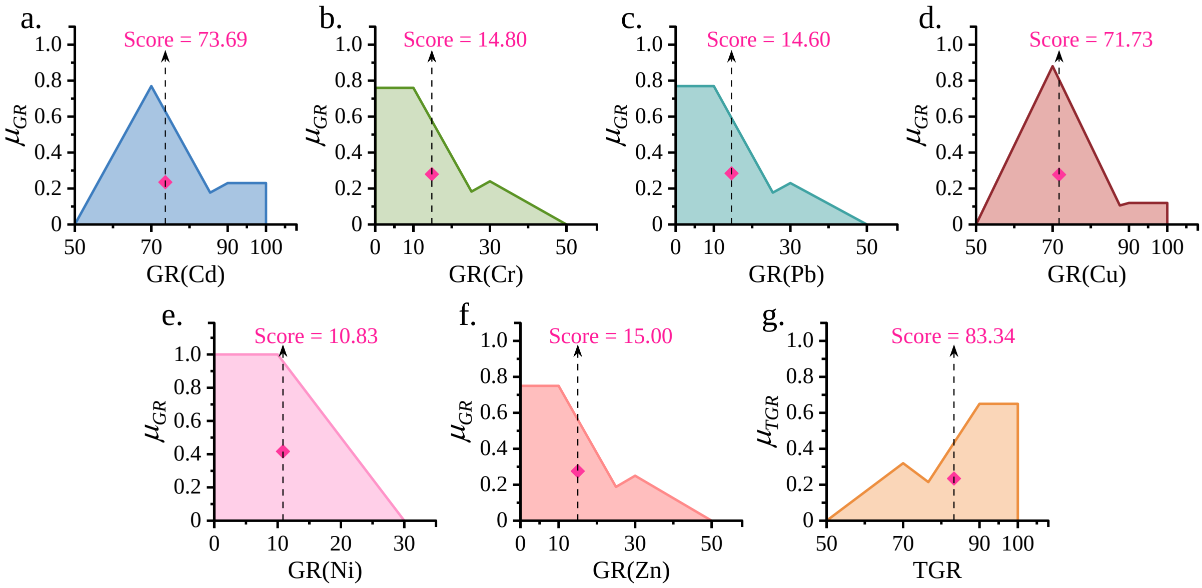

Figure 5. Single-element (**a**–**f**) and multi-element (**g**) integrated general risk fuzzy sets and general risk scores.

In general, the excessive accumulation of Cd and Cu has seriously threatened the overall environmental quality of the study area, resulting in the degradation of farmland and yield reduction. As is second only to these two elements in terms of general risk. As noted in Section 3.2, As is the element with the highest health risk in the study area, particularly in terms of carcinogenic risk. During the soil sampling, we conducted interviews with nearby farmers. Some respondents indicated that congenital diseases and cancers were more common in some villages than others farther away from the mine. This high incidence may be related to the health effects of HMs. Several studies have confirmed that industrial and mining sites are associated with a wide range of diseases in the surrounding population, with emissions of toxic elements significantly increasing their risk of non-carcinogenic and carcinogenic diseases [11,64]. The general risk of the other four elements is lower than that of Cd, Cu, and As. However, the enhanced monitoring of these elements is still needed to prevent new exogenous inputs from increasing accumulation.

4. Conclusions

This work constructed an integrated probabilistic-fuzzy model that sheds new light on soil HM risk assessment and identifying priority pollutants. The model combines probabilistic methods, fuzzy methods, and a logical reasoning system. It can solve the stochastic and fuzzy uncertainty in risk assessments and achieve the joint assessment of environmental and health risks. This study assessed the general risk of soil HMs in a typical Cu mining area in China. The main conclusions are as follows.

We assessed the risk of seven HMs in the soil within the study area from both environmental and health perspectives. Approximately 73.60% of the multi-element total environmental risk and 33.03% of the multi-element total health risk violated their respective risk guidelines. The multi-element general risk score was 83.34, indicating an extremely high-risk level. The single-element general risk (score) decreased in the following order: Cd (73.69) > Cu (71.73) > As (32.86) > Zn (15.00) > Cr (14.80) > Pb (14.60) > Ni (10.83). Cd, Cu, and As are the pollutants of primary concern, while the hazards of the other four HMs on the environment and health are relatively lower. The risks of Cd and Cu are predominantly associated with soil environmental harm, as they are the leading contributors to potential ecological risk (PER) and soil contamination risk (SCR). About 64.55% of the PER of Cd and 62.34% of the SCR of Cu exceeded the thresholds of the environmental guidelines. The risk of As primarily pertains to human health, especially its carcinogenic risk, which requires substantial attention. Due to differences in physiological characteristics (e.g., body weight, skin, etc.) and behavioral habits, children faced higher health risks compared to adults. Approximately 13.58% and 7.41% of the carcinogenic risk posed by As to children and adults, respectively, exceeded the acceptable thresholds stipulated by health guidelines. Overall, comprehensive remediation measures for soil HMP should be implemented in the study area. Furthermore, it is essential to identify the sources of soil HMs and develop source-oriented control strategies to prevent the exacerbation of risks. Cd, Cu, and As are the primary targets for control in the comprehensive management of soil heavy metal(loid) risks in this region. The other four HMs are less hazardous but still require enhanced monitoring.

The complex background of HMP in mining areas usually poses a significant obstacle to risk control. Accurately assessing risks and identifying priority pollutants can help managers develop targeted strategies for improving the efficiency of risk control. The constructed probabilistic-fuzzy model and quantified general risk in this study can provide decision support for managing the risk. However, the membership function in the model may need to be improved through more investigations. In addition, the data employed in this study for risk assessment were the total concentration of HMs in the soil, which may lead to an overestimation of soil heavy metal(loid) risks. In future research endeavors, we will concentrate on the speciation and bioavailability of soil heavy metal(loid) pollutants to provide more precise risk assessment outcomes.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/min13111389/s1, Figure S1: Triangular (a) and trapezoidal (b) fuzzy membership functions. Figure S2: Fuzzy membership function for probabilities of environmental (P_{ER}) and health risks (P_{HR}). Figure S3: General risk score fuzzy membership function. Figure S4: Study area and spatial distribution of soil sampling sites. Table S1: Probability distribution functions of the parameters in health risk assessment model. Table S2: Corresponding reference dose (RfD) and dermal absorption factor (ABS) values of three different exposure pathways for seven heavy metals. Table S3: Corresponding slope factors (SF) of three different exposure pathways for four carcinogenic heavy metal(loid)s. Table S4: Fuzzy rule base for aggregation of fuzzy environmental and health risks into fuzzy general risks. Table S5: Suggestions for risk control of heavy metal(loid) pollution. Table S6: Soil heavy metal(loid) concentration at sampling sites (mg/kg). Text S1: Health risk assessment of heavy metal(loid)s in soils.

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

Funding: This research is funded by the National Natural Science Foundation of China (grant number 41971237) and the National Key Laboratory of Environmental Protection Mining and Metallurgy Resource Utilization and Pollution Control Open Fund (grant number HB201916).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The key research data in the paper has been provided in the Supplementary Materials.

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

Abbreviations

The following abbreviations are used in this manuscript:

HM	Heavy metal(loid)
ER	Environmental risk
HR	Health risk
SCR	Soil contamination risk
PER	Potential ecological risk
CR	Carcinogenic risk
NCR	Non-carcinogenic risk
FER	Fuzzy environmental risk
FHR	Fuzzy health risk
PFGR	Probabilistic-fuzzy general risk
TCR	Multi-element total carcinogenic risk
TNCR	Multi-element total non-carcinogenic risk
P_{ER}	The probability of environmental risk violating the environmental guidelines
P_{HR}	The probability of health risk violating the health guidelines
P_{SCR}	The probability of soil contamination risk violating the environmental guidelines
P_{PER}	The probability of potential ecological risk violating the environmental guidelines
P_{CR}	The probability of carcinogenic risk violating the health guidelines
P_{NCR}	The probability of non-carcinogenic risk violating the health guidelines

References

1. Antoniadis, V.; Shaheen, S.M.; Levizou, E.; Shahid, M.; Niazi, N.K.; Vithanage, M.; Ok, Y.S.; Bolan, N.; Rinklebe, J. A critical prospective analysis of the potential toxicity of trace element regulation limits in soils worldwide: Are they protective concerning health risk assessment?—A review. *Environ. Int.* **2019**, *127*, 819–847. [CrossRef]
2. Binner, H.; Sullivan, T.; Jansen, M.; McNamara, M. Metals in urban soils of Europe: A systematic review. *Sci. Total Environ.* **2023**, *854*, 158734. [CrossRef]
3. Wang, F.; Huo, L.; Li, Y.; Wu, L.; Zhang, Y.; Shi, G.; An, Y. A hybrid framework for delineating the migration route of soil heavy metal pollution by heavy metal similarity calculation and machine learning method. *Sci. Total Environ.* **2023**, *858*, 160065. [CrossRef] [PubMed]
4. Faraji, M.; Alizadeh, I.; Conti, G.O.; Mohammadi, A. Investigation of health and ecological risk attributed to the soil heavy metals in Iran: Systematic review and meta-analysis. *Sci. Total Environ.* **2023**, *857*, 158925. [CrossRef] [PubMed]
5. Zhang, J.; Liu, Z.; Tian, B.; Li, J.; Luo, J.; Wang, X.; Ai, S.; Wang, X. Assessment of soil heavy metal pollution in provinces of China based on different soil types: From normalization to soil quality criteria and ecological risk assessment. *J. Hazard. Mater.* **2023**, *441*, 129891. [CrossRef]
6. Zhang, X.; Zhu, Y.; Li, Z.; Li, J.; Wei, S.; Chen, W.; Ren, D.; Zhang, S. Assessment soil cadmium and copper toxicity on barley growth and the influencing soil properties in subtropical agricultural soils. *Environ. Res.* **2023**, *217*, 114968. [CrossRef]
7. Wu, L.; Yue, W.; Wu, J.; Cao, C.; Liu, H.; Teng, Y. Metal-mining-induced sediment pollution presents a potential ecological risk and threat to human health across China: A meta-analysis. *J. Environ. Manag.* **2023**, *329*, 117058. [CrossRef] [PubMed]
8. Li, H.; Yao, J.; Sunahara, G.; Min, N.; Li, C.; Duran, R. Quantifying ecological and human health risks of metal (loid) s pollution from non-ferrous metal mining and smelting activities in Southwest China. *Sci. Total Environ.* **2023**, *873*, 162364. [CrossRef] [PubMed]
9. Qiao, P.; Wang, S.; Lei, M.; Guo, G.; Yang, J.; Wei, Y.; Gou, Y.; Li, P.; Zhang, Z. Influencing factors identification and the nested structure analysis of heavy metals in soils in entire city and surrounding the multiple pollution sources. *J. Hazard. Mater.* **2023**, *449*, 130961. [CrossRef]
10. Xie, P.; Liu, Z.; Li, J.; Ju, D.; Ding, X.; Wang, Y.; Hower, J.C. Pollution and health-risk assessments of Cr-contaminated soils from a tannery waste lagoon, Hebei, north China: With emphasis on Cr speciation. *Chemosphere* **2023**, *317*, 137908. [CrossRef]
11. Jiang, Y.; Hu, B.; Shi, H.; Yi, L.; Chen, S.; Zhou, Y.; Cheng, J.; Huang, M.; Yu, W.; Shi, Z. Pollution and risk assessment of potentially toxic elements in soils from industrial and mining sites across China. *J. Environ. Manag.* **2023**, *336*, 117672. [CrossRef]
12. Tan, C.; Wang, H.; Yang, Q.; Yuan, L.; Zhang, Y.; Martín, J.D. An integrated approach for quantifying source apportionment and source-oriented health risk of heavy metals in soils near an old industrial area. *Environ. Pollut.* **2023**, *323*, 121271. [CrossRef]
13. Gu, Y.G.; Lin, Q.; Gao, Y.P. Metals in exposed-lawn soils from 18 urban parks and its human health implications in southern China's largest city, Guangzhou. *J. Clean. Prod.* **2016**, *115*, 122–129. [CrossRef]
14. Huang, J.; Wu, Y.; Sun, J.; Li, X.; Geng, X.; Zhao, M.; Sun, T.; Fan, Z. Health risk assessment of heavy metal (loid) s in park soils of the largest megacity in China by using Monte Carlo simulation coupled with Positive matrix factorization model. *J. Hazard. Mater.* **2021**, *415*, 125629. [CrossRef]

15. Yang, S.; Zhao, J.; Chang, S.X.; Collins, C.; Xu, J.; Liu, X. Status assessment and probabilistic health risk modeling of metals accumulation in agriculture soils across China: A synthesis. *Environ. Int.* **2019**, *128*, 165–174. [CrossRef]
16. Ghosh, S.; Banerjee, S.; Prajapati, J.; Mandal, J.; Mukherjee, A.; Bhattacharyya, P. Pollution and health risk assessment of mine tailings contaminated soils in India from toxic elements with statistical approaches. *Chemosphere* **2023**, *324*, 138267. [CrossRef]
17. Zhang, R.; Han, D.; Jiang, L.; Zhong, M.; Liang, J.; Xia, T.; Zhao, Y. Derivation of site-specific remediation goals by incorporating the bioaccessibility of polycyclic aromatic hydrocarbons with the probabilistic analysis method. *J. Hazard. Mater.* **2020**, *384*, 121239. [CrossRef]
18. Gu, X.; Wang, Z.; Wang, J.; Ouyang, W.; Wang, B.; Xin, M.; Lian, M.; Lu, S.; Lin, C.; He, M.; et al. Sources, trophodynamics, contamination and risk assessment of toxic metals in a coastal ecosystem by using a receptor model and Monte Carlo simulation. *J. Hazard. Mater.* **2022**, *424*, 127482. [CrossRef]
19. Zhang, H.; Zhang, F.; Song, J.; Tan, M.L.; Kung, H.-t.; Johnson, V.C. Pollutant source, ecological and human health risks assessment of heavy metals in soils from coal mining areas in Xinjiang, China. *Environ. Res.* **2021**, *202*, 111702. [CrossRef]
20. Chen, H.; Wang, L.; Hu, B.; Xu, J.; Liu, X. Potential driving forces and probabilistic health risks of heavy metal accumulation in the soils from an e-waste area, southeast China. *Chemosphere* **2022**, *289*, 133182. [CrossRef]
21. Mishra, H.; Singh, J.; Karmakar, S.; Kumar, R. An integrated approach for modeling uncertainty in human health risk assessment. *Environ. Sci. Pollut. Res.* **2021**, *28*, 56053–56068. [CrossRef]
22. Zhang, F.; Peng, G.; Xu, P.; Zhu, L.; Li, C.; Wei, N.; Li, D. Ecological risk assessment of marine microplastics using the analytic hierarchy process: A case study in the Yangtze River Estuary and adjacent marine areas. *J. Hazard. Mater.* **2022**, *425*, 127960. [CrossRef]
23. MEEP. *Exposure Factors Handbook of Chinese Population (Adults)*; China Environmental Press: Wuhan, China, 2013.
24. Li, F.; Zhang, J.; Liu, W.; Liu, J.; Huang, J.; Zeng, G. An exploration of an integrated stochastic-fuzzy pollution assessment for heavy metals in urban topsoil based on metal enrichment and bioaccessibility. *Sci. Total Environ.* **2018**, *644*, 649–660. [CrossRef]
25. Chen, X.; Li, F.; Zhang, J.; Liu, S.; Ou, C.; Yan, J.; Sun, T. Status, fuzzy integrated risk assessment, and hierarchical risk management of soil heavy metals across China: A systematic review. *Sci. Total Environ.* **2021**, *785*, 147180. [CrossRef]
26. Guan, Q.; Liu, Z.; Shao, W.; Tian, J.; Luo, H.; Ni, F.; Shan, Y. Probabilistic risk assessment of heavy metals in urban farmland soils of a typical oasis city in northwest China. *Sci. Total Environ.* **2022**, *833*, 155096. [CrossRef]
27. Loh, T.Y.; Brito, M.P.; Bose, N.; Xu, J.; Tenekedjiev, K. Fuzzy system dynamics risk analysis (FuSDRA) of autonomous underwater vehicle operations in the Antarctic. *Risk Anal.* **2020**, *40*, 818–841. [CrossRef]
28. Miao, F.; Zhang, Y.; Li, Y.; Liang, X.; Lin, Q.; Zhou, Y. Establishing a weighted methodology for human health risk assessment of cadmium based on its equilibrium speciation in groundwater. *J. Clean. Prod.* **2021**, *322*, 129053. [CrossRef]
29. Zhao, R.; Guan, Q.; Luo, H.; Lin, J.; Yang, L.; Wang, F.; Pan, N.; Yang, Y. Fuzzy synthetic evaluation and health risk assessment quantification of heavy metals in Zhangye agricultural soil from the perspective of sources. *Sci. Total Environ.* **2019**, *697*, 134126. [CrossRef]
30. Hu, Y.; Wang, Z.; Wen, J.; Li, Y. Stochastic fuzzy environmental risk characterization of uncertainty and variability in risk assessments: A case study of polycyclic aromatic hydrocarbons in soil at a petroleum-contaminated site in China. *J. Hazard. Mater.* **2016**, *316*, 143–150. [CrossRef]
31. Jiang, L.; Sun, H.; Peng, T.; Ding, W.; Liu, B.; Liu, Q. Comprehensive evaluation of environmental availability, pollution level and leaching heavy metals behavior in non-ferrous metal tailings. *J. Environ. Manag.* **2021**, *290*, 112639. [CrossRef]
32. Zerizghi, T.; Guo, Q.; Tian, L.; Wei, R.; Zhao, C. An integrated approach to quantify ecological and human health risks of soil heavy metal contamination around coal mining area. *Sci. Total Environ.* **2022**, *814*, 152653. [CrossRef]
33. Fei, X.; Lou, Z.; Xiao, R.; Ren, Z.; Lv, X. Source analysis and source-oriented risk assessment of heavy metal pollution in agricultural soils of different cultivated land qualities. *J. Clean. Prod.* **2022**, *341*, 130942. [CrossRef]
34. Li, H.; Yao, J.; Min, N.; Duran, R. Comprehensive assessment of environmental and health risks of metal (loid) s pollution from non-ferrous metal mining and smelting activities. *J. Clean. Prod.* **2022**, *375*, 134049. [CrossRef]
35. Zhou, H.; Chen, Y.; Liu, Y.; Wang, Q.; Liang, Y. Farmers' adaptation to heavy metal pollution in farmland in mining areas: The effects of farmers' perceptions, knowledge and characteristics. *J. Clean. Prod.* **2022**, *365*, 132678. [CrossRef]
36. Zhang, Y.; Li, T.; Guo, Z.; Xie, H.; Hu, Z.; Ran, H.; Li, C.; Jiang, Z. Spatial heterogeneity and source apportionment of soil metal (loid) s in an abandoned lead/zinc smelter. *J. Environ. Sci.* **2023**, *127*, 519–529. [CrossRef] [PubMed]
37. MEEP. *China Ecological Environment Bulletin*; Ministry of Ecology and Environment of the People's Republic of China: Beijing, China, 2021.
38. Yang, Q.; Zhang, L.; Wang, H.; Martín, J.D. Bioavailability and health risk of toxic heavy metals (As, Hg, Pb and Cd) in urban soils: A Monte Carlo simulation approach. *Environ. Res.* **2022**, *214*, 113772. [CrossRef] [PubMed]
39. Lei, M.; Li, K.; Guo, G.; Ju, T. Source-specific health risks apportionment of soil potential toxicity elements combining multiple receptor models with Monte Carlo simulation. *Sci. Total Environ.* **2022**, *817*, 152899. [CrossRef]
40. Zhou, H.; Chen, Y.; Yue, X.; Ren, D.; Liu, Y.; Yang, K. Identification and hazard analysis of heavy metal sources in agricultural soils in ancient mining areas: A quantitative method based on the receptor model and risk assessment. *J. Hazard. Mater.* **2023**, *445*, 130528. [CrossRef]
41. MEEP. *Soil Environmental Quality Risk Control Standard for Soil Contamination of Agricultural Land*; Ministry of Ecology and Environment of the People's Republic of China: Beijing, China, 2018.

42. Men, C.; Liu, R.; Xu, L.; Wang, Q.; Guo, L.; Miao, Y.; Shen, Z. Source-specific ecological risk analysis and critical source identification of heavy metals in road dust in Beijing, China. *J. Hazard. Mater.* **2020**, *388*, 121763. [CrossRef]
43. USEPA. *Risk Assessment Guidance for Superfund, Volume I: Human Health Evaluation Manual (Part A)*; Interim Final; Technical Report, EPA/540/1-89/002; Office of Emergency and Remedial Response: Washington, DC, USA, 1989. Available online: https://www.epa.gov/risk/risk-assessment-guidance-superfund-rags-part (accessed on 2 March 2023).
44. Gui, H.; Yang, Q.; Lu, X.; Wang, H.; Gu, Q.; Martín, J.D. Spatial distribution, contamination characteristics and ecological-health risk assessment of toxic heavy metals in soils near a smelting area. *Environ. Res.* **2023**, *222*, 115328. [CrossRef] [PubMed]
45. Yao, Q.; Huang, M.; Zheng, Y.; Chen, M.; Huang, C.; Lin, Q. Prediction and Health Risk Assessment of Copper, Lead, Cadmium, Chromium, and Nickel in Tieguanyin Tea: A Case Study from Fujian, China. *Foods* **2022**, *11*, 1593. [CrossRef]
46. Li, Y.; Ye, Z.; Yu, Y.; Li, Y.; Jiang, J.; Wang, L.; Wang, G.; Zhang, H.; Li, N.; Xie, X.; et al. A combined method for human health risk area identification of heavy metals in urban environments. *J. Hazard. Mater.* **2023**, *449*, 131067. [CrossRef] [PubMed]
47. Liu, Z.; Du, Q.; Guan, Q.; Luo, H.; Shan, Y.; Shao, W. A Monte Carlo simulation-based health risk assessment of heavy metals in soils of an oasis agricultural region in northwest China. *Sci. Total Environ.* **2023**, *857*, 159543. [CrossRef] [PubMed]
48. MEEP. *Technical Guidelines for Risk Assessment of Contaminated Sites*; Ministry of Ecology and Environment of the People's Republic of China: Beijing, China, 2014.
49. Li, T.; Song, Y.; Yuan, X.; Li, J.; Ji, J.; Fu, X.; Zhang, Q.; Guo, S. Incorporating bioaccessibility into human health risk assessment of heavy metals in rice (Oryza sativa L.): A probabilistic-based analysis. *J. Agric. Food Chem.* **2018**, *66*, 5683–5690. [CrossRef]
50. Li, J.; Huang, G.H.; Zeng, G.; Maqsood, I.; Huang, Y. An integrated fuzzy-stochastic modeling approach for risk assessment of groundwater contamination. *J. Environ. Manag.* **2007**, *82*, 173–188. [CrossRef]
51. Hu, G.; Bakhtavar, E.; Hewage, K.; Mohseni, M.; Sadiq, R. Heavy metals risk assessment in drinking water: An integrated probabilistic-fuzzy approach. *J. Environ. Manag.* **2019**, *250*, 109514. [CrossRef] [PubMed]
52. MEEP. *The Technical Specification for Soil Environmental Monitoring*; Ministry of Ecology and Environment of the People's Republic of China: Beijing, China, 2004.
53. MEEP. *Soil and Sediment-Determination of Aqua Regia Extracts of 12 Metal Elements-Inductively Coupled Plasma Mass Spectrometry*; Ministry of Ecology and Environment of the People's Republic of China: Beijing, China, 2016.
54. Yoon, S.; Kim, D.M.; Yu, S.; Park, J.; Yun, S.T. Metal (loid)-specific sources and distribution mechanisms of riverside soil contamination near an abandoned gold mine in Mongolia. *J. Hazard. Mater.* **2023**, *443*, 130294. [CrossRef]
55. Wen, Y.; Li, W.; Yang, Z.; Zhang, Q.; Ji, J. Enrichment and source identification of Cd and other heavy metals in soils with high geochemical background in the karst region, Southwestern China. *Chemosphere* **2020**, *245*, 125620. [CrossRef]
56. Guo, X.; Zhou, T.; Wang, F.; Fan, Y.; Fu, P.; Kong, F. Distribution of Co, Se, Cd, In, Re and other critical metals in sulfide ores from a porphyry-skarn system: A case study of Chengmenshan Cu deposit, Jiangxi, China. *Ore Geology Reviews* **2023**, *158*, 105520. [CrossRef]
57. Yang, S.; He, M.; Zhi, Y.; Chang, S.X.; Gu, B.; Liu, X.; Xu, J. An integrated analysis on source-exposure risk of heavy metals in agricultural soils near intense electronic waste recycling activities. *Environ. Int.* **2019**, *133*, 105239. [CrossRef]
58. Wang, C.C.; Zhang, Q.C.; Kang, S.G.; Li, M.Y.; Zhang, M.Y.; Xu, W.M.; Xiang, P.; Ma, L.Q. Heavy metal (loid) s in agricultural soil from main grain production regions of China: Bioaccessibility and health risks to humans. *Sci. Total Environ.* **2023**, *858*, 159819. [CrossRef] [PubMed]
59. Jiang, H.H.; Cai, L.M.; Wen, H.H.; Hu, G.C.; Chen, L.G.; Luo, J. An integrated approach to quantifying ecological and human health risks from different sources of soil heavy metals. *Sci. Total Environ.* **2020**, *701*, 134466. [CrossRef] [PubMed]
60. Yuan, B.; Cao, H.; Du, P.; Ren, J.; Chen, J.; Zhang, H.; Zhang, Y.; Luo, H. Source-oriented probabilistic health risk assessment of soil potentially toxic elements in a typical mining city. *J. Hazard. Mater.* **2023**, *443*, 130222. [CrossRef] [PubMed]
61. Qu, C.; Shi, W.; Guo, J.; Fang, B.; Wang, S.; Giesy, J.P.; Holm, P.E. China's soil pollution control: Choices and challenges. *Environ. Sci. Technol.* 2016, *50*, 13181–13183. [CrossRef]
62. Sun, J.; Zhao, M.; Huang, J.; Liu, Y.; Wu, Y.; Cai, B.; Han, Z.; Huang, H.; Fan, Z. Determination of priority control factors for the management of soil trace metal (loid) s based on source-oriented health risk assessment. *J. Hazard. Mater.* **2022**, *423*, 127116. [CrossRef]
63. Long, Z.; Zhu, H.; Bing, H.; Tian, X.; Wang, Z.; Wang, X.; Wu, Y. Contamination, sources and health risk of heavy metals in soil and dust from different functional areas in an industrial city of Panzhihua City, Southwest China. *J. Hazard. Mater.* **2021**, *420*, 126638. [CrossRef]
64. Hadzi, G.Y.; Ayoko, G.A.; Essumang, D.K.; Osae, S.K. Contamination impact and human health risk assessment of heavy metals in surface soils from selected major mining areas in Ghana. *Environ. Geochem. Health* **2019**, *41*, 2821–2843. [CrossRef]

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.