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Original Research

Heavy Metal Characterization and Health Risk Assessment in Agricultural Soils from an Agate Dyeing Village

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Abstract

The pollution of heavy metals in agricultural soil poses a dangerous health risk to nearby residents. This study presents a method to assess the health risks of heavy metal pollution using Monte Carlo simulations. Soil samples from an agate dyeing village were collected and their heavy metal contents were analyzed. The results indicate the contamination of heavy metals in the study area soil, the single-factor index values of Pb, Cu, Cr, and Ni were found to indicate mild pollution, and the Nemerow complex index values of all investigated metals were ranked at level IV, indicating moderate pollution. The health assessment indicates an unacceptable total carcinogenic risk for adults and children and non-carcinogenic hazard quotient for adults. Oral ingestion was found to be the main exposure pathway, and the most sensitive factors were the adherence factor, soil ingestion rate, and body weight.

Keywords: agricultural soil, heavy metal, agate dyeing, health risk assessment, Monte Carlo simulation

Introduction

Metals and metalloids with densities higher than 5 g/cm^3 are referred to as heavy metals [1] and can pose serious health risks when excessively consumed by humans and animals. The concentrations of heavy metals in agricultural soil have been shown to reach high pollution levels due to the influence of anthropogenic activities (e.g., mining, fossil fuel combustion, sewage irrigation, metal smelting, fertilizer and pesticide application, traffic source discharge) [2-4]. High heavy metal concentrations in soil not only reduce the environmental quality but also pose serious health risks to nearby residents and other organisms by entering the food chain [5]. For example, acute and chronic arsenic exposure can lead to cardiovascular disorders and other maladies that may ultimately lead to cancer [6-8]. A comprehensive understanding of the sources and pollution characteristics of heavy metals

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in agricultural soil is therefore of great importance to protect human health and the environment [9, 10].

Rapid social and economic development in China has led to increasingly severe soil heavy metal pollution conditions and environmental problems, especially in agricultural regions [7, 11]. A national soil pollution survey conducted in 2014 by the Ministry of Environmental Protection of China and the Ministry of Land and Resources [12] reported that soils in some regions have been heavily contaminated, and that the cultivated land soil quality is particularly alarming. For example, a considerable proportion of soils in agricultural regions in China have been contaminated by heavy metals, with at least 19.4% of arable land being classified as above standard pollution levels [7, 13], thus leading to a diminishing availability of arable lands. Chinese officials have therefore implemented strong measures to mitigate and prevent soil pollution to improve soil quality, protect the health of humans and other life forms, and ensure the quality of agricultural products. For instance, a national action plan "Soil Ten Chapter" was implemented to prevent and control the pollution of heavy metals in soil. The "Soil Ten Chapter" clarifies the overall idea of soil pollution control and prevention in China, which is to reduce the current extent of land pollution and restore contaminated land. Its main prevention and control measures include the establishment of a sound soil pollution prevention and legal control system to better understand and improve the status quo of soil environmental quality, classification and grading management, and strict monitoring of existing pollution sources to achieve the aim of initial prevention followed by combined prevention and control.

Previous studies of heavy metal contamination and associated health risk assessments involving agricultural soils were mostly concentrated near smelting- and mining-impacted areas [14-18], whereas the influence of the agate dyeing industry on soil pollution has not been reported. In terms of health risk estimations, most previous studies have typically performed a deterministic risk quantification of the exposure to heavy metal-contaminated soils on the regional scale [19, 20]. However, deterministic risk assessments mainly consider the total heavy metal concentrations and most probable exposure parameters, which may either over- or under-estimate the risk [21-23]. The most hazardous elements for a population are also difficult to discern using deterministic methods owing to the heavy metal concentration uncertainties and specific variabilities among individuals [14-18].

A recent review of heavy metal concentrations in Chinese agricultural soils addressed six heavy metals in terms of pollution assessment and risk screening, including cadmium (Cd), arsenic (As), nickel (Ni), chromium (Cr), zinc (Zn), and lead (Pb) [11]. Monte Carlo simulations have been performed to determine the probabilistic health risks, associated uncertainties, and variations in the toxicity parameters, body weight, and ingestion rates. The results showed that As had both the highest cancer and non-cancer risks, while Cd and Cr metals also posed substantial cancer risks. Sensitivity analyses showed that soil ingestion rates and metal concentrations are the predominant contributors to the total risk variance. Another review addressed the deterministic health risks associated with soil ingestion exposure to eight heavy metals in 72 mining-impacted areas in China, and concluded that Cd, As, Pb, and Ni are the dominant contributors to the total local residents' non-cancer risks [24]. Monte Carlo simulations are the most commonly applied method for probabilistic risk analysis and can be used to determine the risk probability of exceeding a particular guideline value [14] and to identify the particular priority metal for risk control [11, 25].

To address the problems highlighted above, this study's objectives are to: 1) explore the condition of heavy metal contamination in agricultural soils near the agate dyeing industry; 2) determine the probabilistic risks of exposure to heavy metals in soils of the study area considering both the variability and uncertainty of the key exposure parameters; and 3) perform a probabilistic risk-based ranking of heavy metals and a sensitivity analysis of the exposure factors. Samples were collected in an area known for heavy metal contamination owing to the agate dyeing industry, and the single-factor and Nemerow complex pollution indices were applied to assess the heavy metal contamination factors. Monte Carlo simulations were performed to assess the carcinogenic and non-carcinogenic risks. The results obtained herein provide an important reference for developing remediation strategies to minimize heavy metal exposure and protect human health [26].

Material and Methods

Study Area

Shijiazi Town is located 48 km east of the county seat of Fuxin Mongolian Autonomous County, Liaoning Province, and is home to China's largest professional agate market, which has developed over the past half century. A statistical study from 2005 reported more than 3000 households in the town with more than 8000 people employed in the agate processing industry.

Since the 1970s, agate processing has been shown to pollute groundwater and soil owing to the dumping of agate dyeing wastewater into river ditches. The main compounds used in the agate dyeing industry include zinc sulfate, nickel sulfate, and copper oxide powder. Dyeing farms use approximately 110 tons of these compounds on an annual basis and produce more than 20,000 tons of wastewater. Disorderly production and random wastewater dumping have led to varying degrees of local groundwater and soil pollution.

Nalishan Village presently suffers from the most serious degree of pollution related to the agate dyeing industry, and was therefore selected herein as a case study. Data from a 2006 study of the Environmental Monitoring Station of Fuxin Mongolian Autonomous County indicated that heavy metals in the groundwater of Shijiazi town exceed the guideline values and that dyeing wastewater leakage poses a serious problem; however, the degree of soil pollution in the area remains unclear.

Sampling and Analysis

Soil samples were collected from a sewage ditch in Nalishan Village in strips according to the flow direction with intervals of approximately 30 m, which were adjusted according to the topographic characteristics and landforms. The sampling point density increased near the agate farms (Fig. 1). Samples were collected between August 6 and 15, 2020 from soil depths of 0-0.2 m at 110 sampling sites.

For Cd, Pb, Cu, Cr, Zn, and Ni, the samples were digested using $HF/HNO_3/HClO_4$, and analyzed by atomic absorption spectrophotometry (Varian, Spectr AA 220, American) [27]. For Hg and As, the samples were digested with $V_2O_5/H_2SO_4/HNO_3$, then, Hg, and As were determined using cold vapor atomic fluorescence spectrometry (Tekran 2500, CVAFS). Each sample was tested three times. Standard solutions and reagent blanks were randomly inserted for precision analysis and quality control and recovery rates ranged from 90.2% to 105.3% [28]. The reagents used in the experiment were all high grade. The glassware was soaked in 3 mol/L nitric acid for 24 h prior to use, rinsed with tap water, and then rinsed three times with secondary deionized water [29].

Soil Contamination Assessment

Single-Factor Pollution Index

Quantitative and qualitative methods were applied to determine the anthropogenic metal contamination intensity in the soil samples. The qualitative single-factor pollution index method was used in this study [30, 31]. The contamination factor P_i was calculated according to:

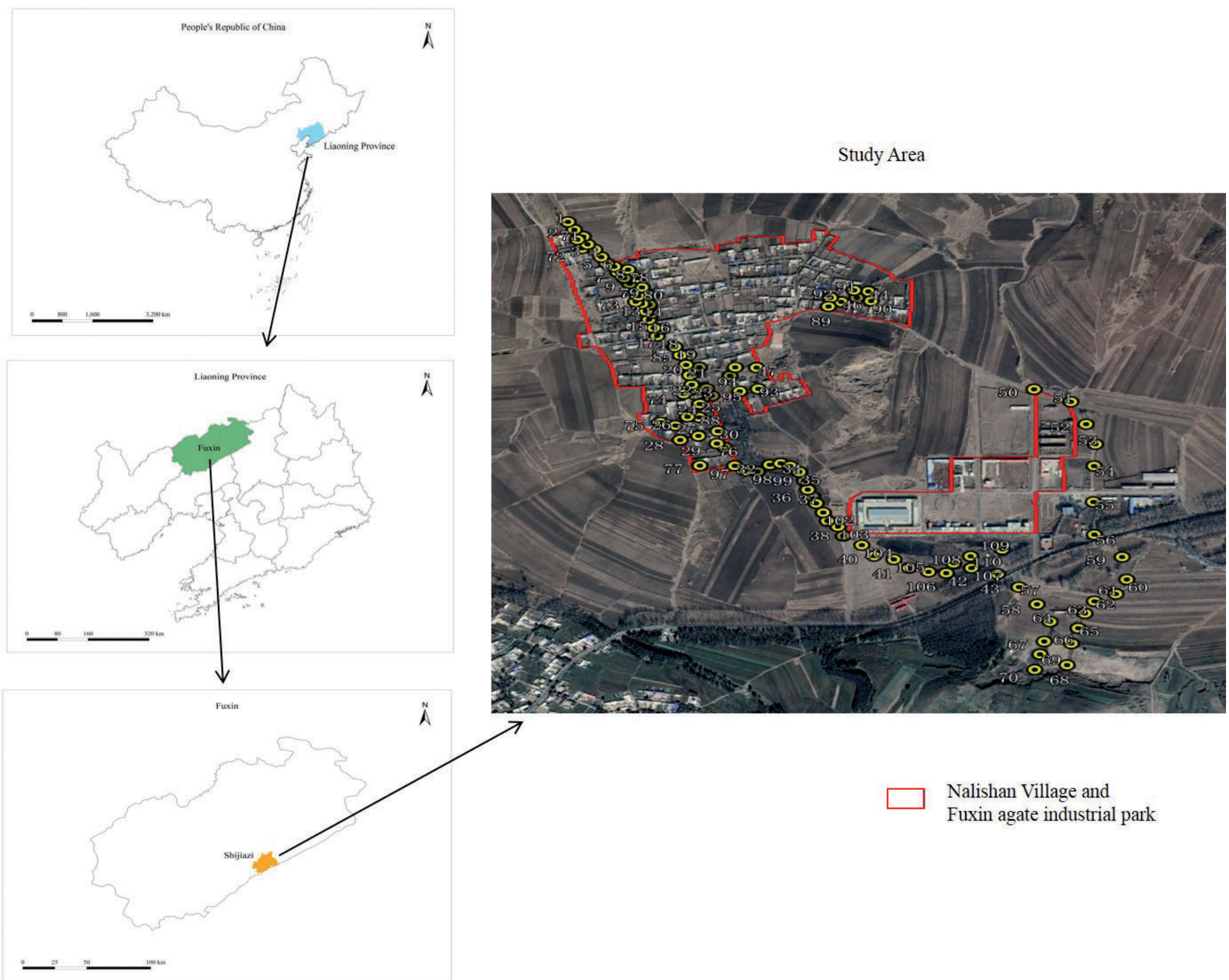

Fig. 1. Study area location and sampling site distribution.

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

where C_i is the concentration of metal *i* at a given site, and S_i is the background concentration of the same metal at a reference site. When $P_i \leq 1$, the surface soil is free of pollution; when $P_i > 1$, the background value of the heavy metals in the surface soil has been artificially enhanced. Larger P_i values are associated with more serious surface soil pollution.

Complex Pollution Index

Nemerow complex pollution indices were applied to fully assess the soil samples according to:

$$P_N = \sqrt{\frac{(P_{\max})^2 + (P_{ave})^2}{2}} \tag{2}$$

where P_N, $P_{\max}$, and P_{ave} represent the contamination factor for an individual element, maximum contamination factor, and average contamination factor, respectively. The evaluation criteria for both pollution indices are listed in Table S1.

Health Risk Assessment

The main aim of a health risk assessment is to establish a quantitative relationship between the degree of human health and environmental pollution, and to estimate the probability and degree of human health damage that may be driven by environmental pollution as a measure to determine the priority control of pollutants [32]. In 1983, the U.S. Environmental Protection Agency proposed a "four-step method" for health risk assessment, including exposure assessment, hazard identification, risk characterization, and toxicity assessment [33].

The study area is a village and thus, the exposed population includes both adults and children. Adults can be exposed to soil via two pathways, namely dermal contact and oral ingestion [34]. The average daily intake of heavy metals can be applied to calculate the exposure dose following the procedure of the United States Environmental Protection Agency [35, 36]. The average daily heavy metal intake via the oral ingestion pathway is given as:

$$CDI_I = \frac{CS \times SIR \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{3}$$

where *CS* is the heavy metal concentration in the soil (mg/kg), CDI_I is the chronic daily intake from soil oral ingestion (mg/kg·day), *SIR* is the soil ingestion rate, *ED* and *EF* are the exposure duration and frequency, respectively, *AT* is the average exposure time, and *BW* is body weight. The average daily heavy metal intake via the dermal contact pathway is given as:

$$CDI_D = \frac{CS \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{4}$$

where CDI_D is the chronic daily intake from dermal contact (mg/kg·day), *AF* is the adherence factor, *ABS* is the dermal absorption factor, and *SA* is the exposed skin surface area.

Table 1 [37-43] presents detailed information on the probabilistic exposure factors. Exposure assessments were determined by performing Monte Carlo simulations using Crystal Ball 11.1 software with the listed parameters. The model was run for 10,000 iterations and the confidence level was determined to be 95%, from which an approximate risk evaluation value was obtained.

Carcinogenic Risk Assessment

Carcinogenic risk is estimated as the incremental probability for which an individual cancer will develop over a lifetime owing to potential carcinogen exposure [44]. This risk is described as follows:

$$CR = CDI \times SF \tag{5}$$

where *SF* is the carcinogenicity slope factor (mg/kg·day) and *CR* is a unit-less probability of an individual developing cancer over their lifetime.

The sum of the average contribution of individual heavy metals for all considered pathways is given as the total excess lifetime cancer risk:

$$CR_{total} = \sum CR_i \tag{6}$$

where CR_{total} represents the risk contributions through dermal contact and oral ingestion pathways.

Non-carcinogenic Risk Assessment

The hazard quotient (*HQ*) is frequently applied or characterized for non-carcinogenic hazards. The *HQ* is defined as the average daily intake of the toxicity threshold value of a specific heavy metal, which is called the chronic reference dose (*RfD*) (mg/kg·day). For a single chemical, the *HQ* is given as:

$$HQ = \frac{CDI}{RfD} \tag{7}$$

Non-carcinogenic effects are defined by the sum of all *HQ* values when considering more than one chemical, which is referred to as the hazard index (HQ_{total}) and calculated according to:

Table 1. Monte Carlo parameter distribution. Superscripts 1 and 2 indicate adults and children, respectively.

Parameter	Symbol	Type	Distribution	Units
Body weight[1]	BW_a	Log-normal	58.7±12.0	kg
Body weight[2]	BW_c	Log-normal	37.3±9.1	kg
Average exposure time[1]	AT_a	Point	25,550 (Carcinogenic compound)	day
			8760 (Non-carcinogenic compound)	
Average exposure time[2]	AT_c	Point	25,550 (Carcinogenic compound)	day
			2190 (Non-carcinogenic compound)	
Exposure duration[1]	ED_a	Point	70	year
Exposure duration[2]	ED_c	Point	18	year
Exposure frequency[1]	EF_a	Log-normal	252±1.01	day/year
Exposure frequency[2]	EF_c	Log-normal	252±1.01	day/year
Exposed skin surface area[1]	SA_a	Triangular	1530 (760–4220)	cm^2
Exposed skin surface area[2]	SA_c	Triangular	860 (430–2160)	cm^2
Soil ingestion rate[1]	SIR_a	Triangular	25(0.1–50)	mg/day
Soil ingestion rate[2]	SIR_c	Log-normal	24±4	mg/day
Adherence factor[1]	AF_a	Log-normal	0.49±0.54	$mg/cm^2 \cdot day$
Adherence factor[2]	AF_c	Log-normal	0.65±1.2	$mg/cm^2 \cdot day$
Dermal absorption factor (Cd)	ABS_{Cd}	Point	0.14	Unitless
Dermal contact factor (Hg)	ABS_{Hg}	Point	0.10	Unitless
Dermal contact factor (As)	ABS_{As}	Point	0.95	Unitless
Dermal contact factor (Pb)	ABS_{Pb}	Point	0.50	Unitless
Dermal contact factor (Cr)	ABS_{Cr}	Point	0.04	Unitless
Dermal contact factor (Cu)	ABS_{Cu}	Point	0.56	Unitless
Dermal contact factor (Ni)	ABS_{Ni}	Point	0.04	Unitless
Dermal contact factor (Zn)	ABS_{Zn}	Point	0.25	Unitless

$$HQ_{total} = \sum HQ_i \tag{8}$$

The *SF* and *RfD* values are listed in Table S2, in which the risk determines the carcinogenic risk and *HQ* of the pollutant. An unacceptable risk is assigned if the carcinogenic risk value exceeds 10^{-6} or if *HQ* exceeds 1. Several evaluation grading standards are combined to comprehensively determine the health risk degree. The health hazards are then divided into five levels according to the evaluation standards defined in Table S3.

Results and Discussion

Heavy Metal Pollution Characteristics of the Soil Samples

Fig. 2 and Table 2 demonstrate that the Cd, Cu, Ni, and Cr contents exceeded their individual risk screening values for the contamination of agricultural soil in China (GB15618-2018) at 1, 5, 2, and 3 different sampling sites, respectively, yielding excesses of approximately 0.91%, 4.55%, 1.82%, and 2.73% above the latest risk screening values. The single-factor index values of Pb, Cr, Ni, and Cu were found to be greater than 1 (Table S4), which implies mild pollution, whereas those of Cd, Hg, As, and Zn were lower than 1, indicating safety. Among these, the excess degree ranks as follows: Cu>Pb>Ni>Cr. Cd is classified as safe due to its low detection rate, even though its concentration exceeds the standard value at a few sampling sites (Fig. 2). The Nemerow complex index values of the eight heavy metals all exceeded 2 (level IV), indicating moderate pollution [15].

The heavy metal content in the study area was found to be lower than that in other areas (Table 3), similar to data reported for farmland soil in northwestern Xiushan, Chongqing. The contents of Cd, Hg, Cr, and Zn were significantly lower than those in other regions. Because the sampling site is located around a seasonal

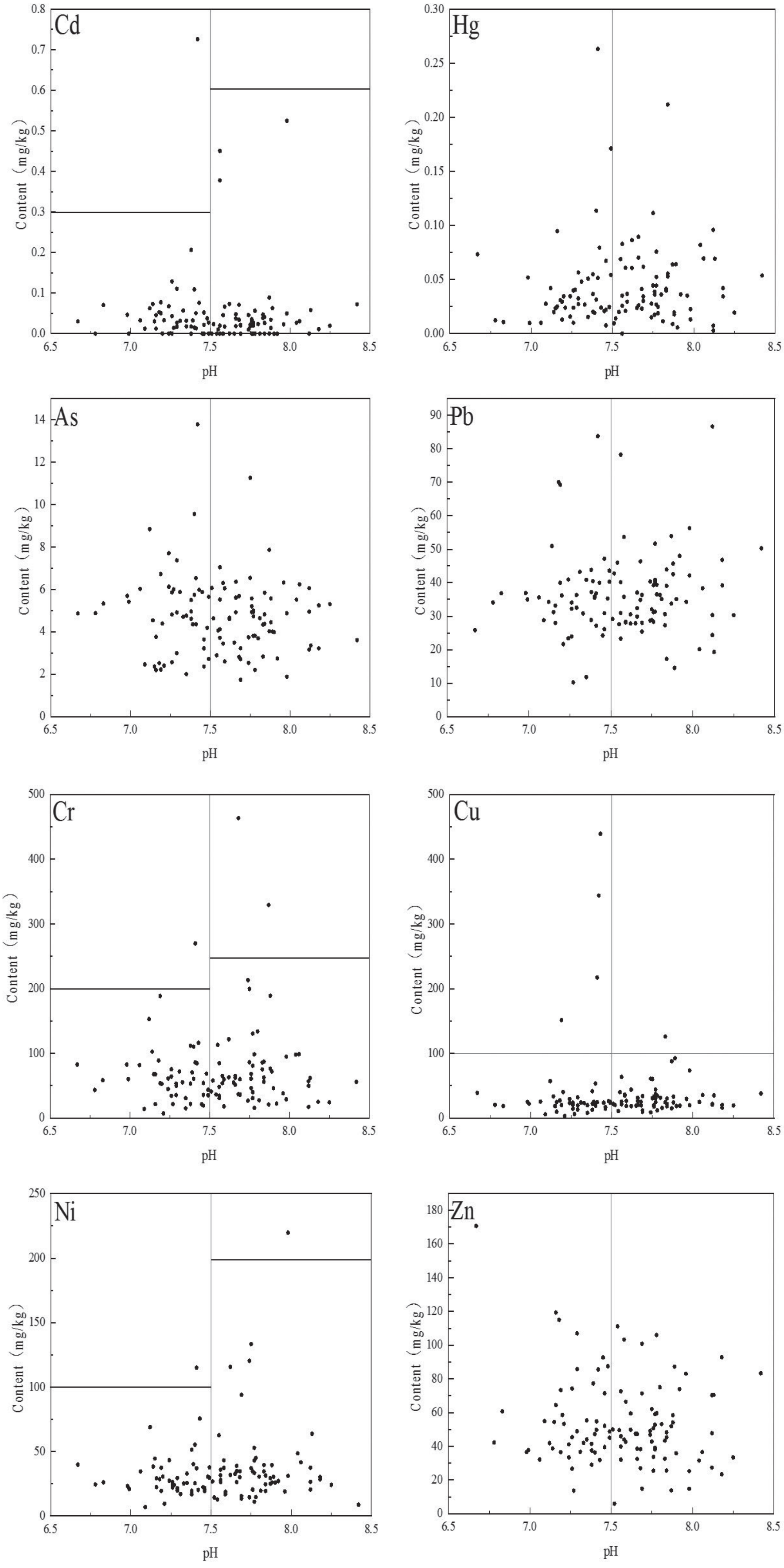

Fig. 2. Heavy metal concentrations measured in the soil samples as a function of pH. The horizontal lines indicate the standard value over the corresponding pH interval. Panels without a horizontal line indicate that the metal contents are substantially below the standard value.

Table 2. Statistical summary of the concentrations of heavy metals in the study area.

Elements	Cd	Hg	As	Pb	Cu	Cr	Zn	Ni
Min (mg/kg)	0	2.3×10^{-6}	1.74	10.21	3.38	7.23	5.97	6.85
Median (mg/kg)	0.03	0.03	4.72	35.11	23.84	58.15	47.60	28.88
Max (mg/kg)	0.73	0.26	13.78	86.67	439.09	463.38	170.71	219.82
Mean (mg/kg)	0.05	0.04	4.75	36.44	37.15	71.36	53.36	35.28
Standard deviation (mg/kg)	0.10	0.04	1.87	12.51	56.19	62.94	25.88	28.14
Coefficient of variation (%)	205.80	89.30	39.25	34.34	151.26	88.20	48.51	79.76
Positive rate (%)	75.45	100	100	100	100	100	100	100
Exceeding standard rate (%)	0.91	0	0	0	4.55	2.73	0	1.82

Table 3. Content characteristics of heavy metals in the study area compared with other regions (mg/kg).

Location	Cd	Hg	As	Pb	Cr	Cu	Ni	Zn	References
Study Area	0.05	0.04	4.75	36.44	37.15	71.36	53.36	35.28	This Study
Qixia Mining Area, China	3.66	-	92.78	637.15	86.58	-	-	684.91	[45]
Xiushan, Chongqing, China	0.59	1.18	18.81	41.86	82.34	36.33	36.12	112.61	[46]
Jiangxi Tungsten Mine, China	2.03	0.65	61.04	98.74	64.64	126	91.38	-	[47]
Sinop Province, Turkey	-	-	5.66	17.01	194.73	43.19	85.02	65.11	[48]

river in the village and not within a cultivated area, the source of other heavy metal pollution is not directly clear and its pollution degree is not high.

Risk Characterization Based on Monte Carlo Simulations

The carcinogenic health risks determined by the Monte Carlo simulations are presented in Table 4. For adults and children, the 95% quantile carcinogenic risk values of As and Cr exceed 10^{-6}, belonging to grades II and I (i.e., low-middle and low risks), respectively, and the risk value is slightly higher for adults than for children. The total carcinogenic risk for both adults and children exceeds 10^{-6} and is classified as grades III and II, respectively, which are middle and low-middle risks. The soil intake rate parameter is found to be considerably larger for the oral ingestion pathway than the exposed skin surface area and adherence factor for the dermal contact pathway, and leads to a more direct exposure to pollutants in the soil. Oral ingestion is thus the main exposure route.

Table 5 shows that the 95% quantile non-carcinogenic *HQ* value of the eight investigated heavy metals and total non-carcinogenic *HQ* values were lower than 1 for children, which falls within an acceptable risk. The 95% quantile non-carcinogenic *HQ* values of the eight heavy metals were also lower than 1 for adults; however, the total non-carcinogenic *HQ* exceeded 1, thus indicating an unacceptable risk. The calculation of non-carcinogenic risk involves differences in the parameters of body weight and exposure time between adults and children, thus resulting in higher risk values for adults than children. Oral ingestion was also the main exposure pathway.

Fig. 3 shows the contributions of the different heavy metals to the CR_{total} and HQ_{total} values for adults and children. The results indicate that Cr makes the highest contribution to CR_{total} with a similar contribution rate for children and adults. Pb and As are found to make the highest contributions to HQ_{total} for adults and children.

Sensitivity Analysis

Sensitivity analysis was conducted based on the Monte Carlo probability model to explore the impact of each exposure parameter on the health risk, and the results are shown in Fig. 4. Higher absolute sensitivity values indicate a higher impact on the risk value. For CR_{total}, *AF* and *SIR* were the most sensitive factors for both adults and children, yielding 65.11% and 56.21%, respectively. For HQ_{total}, *AF* and *BW* were the most sensitive factors for both adults and children, yielding 59.23% and 61.81%, respectively.

Positive sensitivity values indicate a positive correlation with the risk evaluation value, whereas negative values indicate a negative correlation. For CR_{total} and HQ_{total} for both adults and children, *AF* is found to have the strongest positive correlation, whereas *BW* for adults has the strongest negative correlation.

Table 4. Results of carcinogenic risks based on the Monte Carlo simulations.

Heavy Metal	Median				SD				95%					
	Adults		Children		Adults		Children		Adults			Children		
	Ingestion	Dermal	Ingestion	Dermal	Ingestion	Dermal	Ingestion	Dermal	Ingestion	Dermal	Sum	Ingestion	Dermal	Sum
Cd	8.84×10^{-8}	3.20×10^{-8}	3.46×10^{-8}	6.85×10^{-9}	6.51×10^{-8}	4.18×10^{-8}	3.87×10^{-8}	1.10×10^{-8}	1.65×10^{-7}	1.62×10^{-7}	3.27×10^{-7}	5.94×10^{-8}	5.61×10^{-8}	1.15×10^{-7}
As	2.12×10^{-6}	1.27×10^{-6}	8.30×10^{-7}	2.72×10^{-7}	2.58×10^{-6}	1.00×10^{-6}	1.53×10^{-6}	2.63×10^{-7}	6.44×10^{-6}	3.95×10^{-6}	1.04×10^{-5}	2.35×10^{-6}	1.34×10^{-6}	3.70×10^{-6}
Cr	1.06×10^{-5}	4.39×10^{-6}	4.15×1^{0}-6	9.39×10^{-7}	8.91×10^{-6}	5.01×10^{-6}	5.30×10^{-6}	1.31×10^{-6}	2.22×10^{-5}	1.98×10^{-5}	4.20×10^{-5}	8.13×10^{-6}	6.73×10^{-6}	1.49×10^{-5}
CR_{total}									2.88×10^{-5}	2.39×10^{-5}	5.27×10^{-5}	1.05×10^{-5}	8.13×10^{-6}	1.87×10^{-5}
									54.72%	45.28%		56.47%	43.53%	

Table 5. Results of non-carcinogenic hazard quotients based on the Monte Carlo simulations.

Heavy Metal	Median				SD				95%					
	Children		Adults		Children		Adults		Children			Adults		
	Ingestion	Dermal	Ingestion	Dermal	Ingestion	Dermal	Ingestion	Dermal	Ingestion	Dermal	Sum	Ingestion	Dermal	Sum
Hg	1.92×10^{-4}	2.58×10^{-5}	1.22×10^{-3}	3.20×10^{-4}	6.07×10^{-5}	1.10×10^{-4}	5.92×10^{-4}	6.21×10^{-4}	3.14×10^{-4}	2.12×10^{-4}	5.26×10^{-4}	2.33×10^{-3}	1.63×10^{-3}	3.95×10^{-3}
Cd	6.57×10^{-4}	2.95×10^{-5}	4.18×10^{-3}	3.47×10^{-4}	2.07×10^{-4}	1.26×10^{-4}	2.02×10^{-3}	6.75×10^{-4}	1.07×10^{-3}	2.43×10^{-4}	1.31×10^{-3}	7.94×10^{-3}	1.77×10^{-3}	9.71×10^{-3}
Pb	1.40×10^{-2}	2.67×10^{-3}	7.76×10^{-2}	8.91×10^{-2}	4.42×10^{-3}	5.19×10^{-3}	1.51×10^{-1}	4.31×10^{-2}	2.29×10^{-2}	1.37×10^{-2}	3.66×10^{-2}	3.95×10^{-1}	1.69×10^{-1}	5.64×10^{-1}
As	2.13×10^{-2}	3.01×10^{-3}	1.36×10^{-1}	3.54×10^{-2}	6.73×10^{-3}	1.29×10^{-2}	6.57×10^{-2}	6.89×10^{-2}	3.48×10^{-2}	2.48×10^{-2}	5.96×10^{-2}	2.58×10^{-1}	1.80×10^{-1}	4.38×10^{-1}
Ni	4.97×10^{-4}	2.37×10^{-3}	1.51×10^{-2}	5.84×10^{-4}	2.12×10^{-3}	7.49×10^{-4}	7.31×10^{-3}	1.14×10^{-3}	4.09×10^{-3}	3.87×10^{-3}	7.96×10^{-3}	2.87×10^{-2}	2.97×10^{-3}	3.17×10^{-2}
Cr	2.78×10^{-5}	6.40×10^{-5}	3.27×10^{-4}	4.07×10^{-4}	1.19×10^{-4}	2.02×10^{-5}	6.36×10^{-4}	1.97×10^{-4}	2.29×10^{-4}	1.04×10^{-4}	3.33×10^{-4}	1.67×10^{-3}	7.74×10^{-4}	2.44×10^{-3}
Cu	3.29×10^{-4}	1.25×10^{-3}	3.88×10^{-3}	7.95×10^{-3}	1.41×10^{-3}	3.94×10^{-4}	7.54×10^{-3}	3.85×10^{-3}	2.71×10^{-3}	2.04×10^{-3}	4.75×10^{-3}	1.97×10^{-2}	1.51×10^{-2}	3.48×10^{-2}
Zn	2.39×10^{-4}	3.38×10^{-5}	1.52×10^{-3}	3.98×10^{-4}	7.55×10^{-5}	1.44×10^{-4}	7.37×10^{-4}	7.73×10^{-4}	3.90×10^{-4}	2.78×10^{-4}	6.69×10^{-4}	2.90×10^{-3}	2.02×10^{-3}	4.92×10^{-3}
HQ_{total}									6.64×10^{-2}	4.53×10^{-2}	1.12×10^{-1}	7.16×10^{-1}	3.74×10^{-1}	1.09
									59.47%	40.53%		65.69%	34.31%	

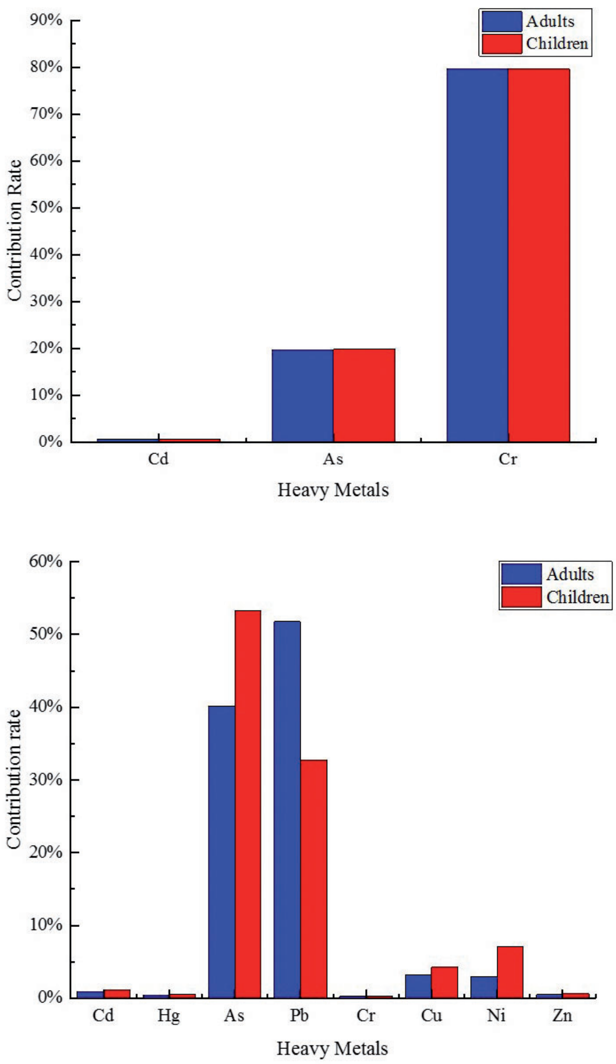

Fig. 3. Contribution rate of CRtotal (upper panel) and HQtotal (lower panel).

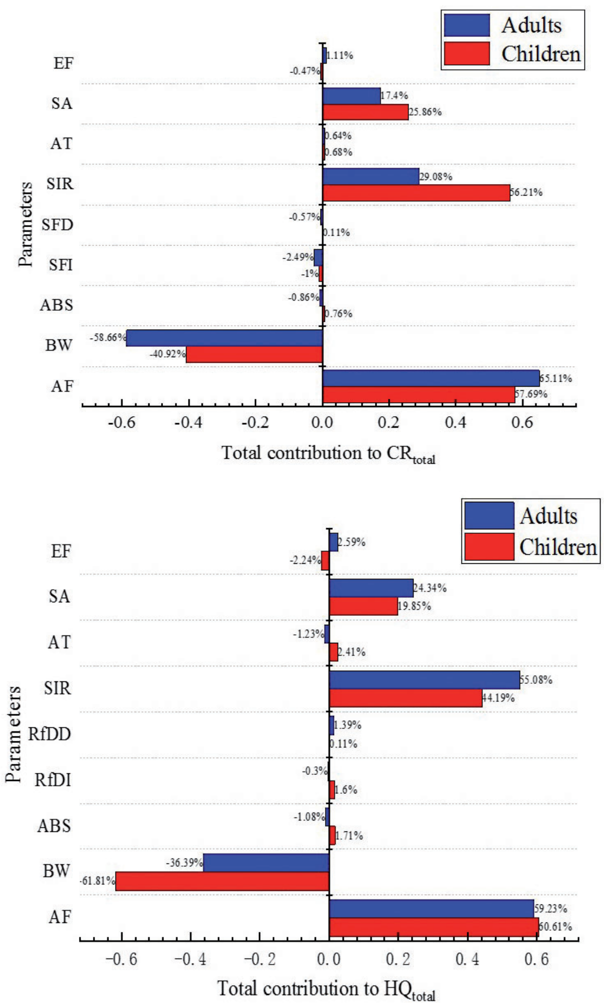

Fig. 4. Sensitivity analysis of CRtotal (upper panel) and HQtotal (lower panel).

Control Strategy

Prevention at the source is fundamental. A number of systems must be established, such as a soil environmental impact assessment, soil toxic and harmful substance lists, soil pollution key supervision unit lists, and soil pollution prevention and control emergency plans to comprehensively eliminate soil pollution sources, reduce soil pollution input, and strictly prevent new soil pollution risks.

Process control is the focus. The implementation of process control, pollution investigation, pollution monitoring, and emergency response are recommended as management principles.

System governance is the key. There is an urgent need to speed up the research of key risk management and control technologies and promotion of large-scale engineering applications to give full play to their sustainability, flexibility, and systemic advantages.

Management. Intelligent management is critical. The priority of risk zoning and management must be determined to improve soil pollution monitoring and early risk warning capabilities.

Conclusions

This study applied the Monte Carlo simulation technique to determine the health risks associated with exposure to soil polluted with heavy metals in an agate dyeing village. The health risk was investigated for all potentially exposed local residents. The specific conclusions are summarized as follows:

The dyes involved in the agate production process contain heavy metals, and the study area is already polluted with heavy metals. The Cd, Cu, Ni, and Cr contents in the study area exceed their standard values. The single-factor index values of Pb, Cr, Ni, and Cu were found to be greater than 1, which implies mild pollution. The Nemerow complex index values of the eight heavy metals all exceeded 2 (level IV), indicating moderate pollution. The main reason is that the villagers actively dye agate and dump waste water containing heavy metal compounds at will, resulting in soil pollution.

The results of the health risk assessment based on the Monte Carlo simulation show that the total carcinogenic risk for adults and children and the non-carcinogenic hazard quotients for adults are higher than the standard values. Oral ingestion is the main route of exposure. In particular, the contribution rates of Cr, As, and Pb are excessively high. The sensitivity analysis shows that the *AF* (adherence factor), *BW* (body weight), and *SIR* (soil ingestion rate) are the most sensitive factors. Body weight is negatively sensitive, which indicates that individuals with a lower weight face a more serious effect.

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Conflict of Interest

The authors declare no conflict of interest.

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Supplementary Tables

Table S1. Evaluation criteria of heavy metal pollution indices for soil.

Class	Nemerow complex pollution index method		Single-factor index method	
	Value	Level	Value	Level
1	$P_N < 0.7$	I (Safety)	$P_i \leq 1$	Safety
2	$0.7 < P_N < 1.0$	II (Warning line)	$1 < P_i \leq 2$	Mild pollution
3	$1.0 < P_N < 2.0$	III (Mild pollution)	$2 < P_i \leq 3$	Middle pollution
4	$2.0 < P_N < 3.0$	IV (Middle pollution)	$3 < P_i$	Severe pollution
5	$3.0 < P_N$	V (Severe pollution)		

Table S2. Toxicological data for heavy metals. Subscripts *D* and *I* represent dermal contact and oral ingestion, respectively (Qu et al. 2012).

	SF_I per (mg/kg·day)	SF_D per (mg/kg·day)	RfD_I (mg/kg·day)	RfD_D (mg/kg·day)
Cd	6.1	6.1	1×10^{-4}	4.4×10^{-5}
Hg			3×10^{-4}	3×10^{-5}
As	1.5	3.66	3×10^{-4}	2.85×10^{-4}
Pb			3.5×10^{-3}	5.25×10^{-4}
Cr	0.5	20	1.5	0.0195
Cu			0.04	0.012
Ni			0.02	0.0054
Zn			0.3	0.075

Table S3. Classification of health risk standards.

Level	Range	Risk
I	1×10^{-6}–1×10^{-5}	low
II	1×10^{-5}–5×10^{-5}	low-middle
III	5×10^{-5}–1×10^{-4}	middle
IV	1×10^{-4}–5×10^{-4}	middle-high
V	5×10^{-4}–1×10^{-3}	High

Table S4. Results of the soil pollution assessment.

P_i								P_N	Level
Hg	Cd	Pb	As	Cu	Cr	Zn	Ni		
1.19	0.44	1.69	0.52	1.70	1.34	0.97	1.36	2.12	IV