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Eco-geochemical evaluation of the Leizhou Peninsula (southern China) and the prediction of heavy metal content in soils

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ABSTRACT

The Leizhou Peninsula is an important base for tropical and subtropical cash crops in China, but still lacks systematic research on regional eco-geochemical characteristics. Here the elemental results show that risk-free soils accounted for 9168 km^2 and were mainly concentrated in the northern Leizhou Peninsula, while risk-controllable soils occupied 3318 km^2 and were mostly distributed in the southern part. The contributor of the heavy metals in soils was mainly natural rocks, while the road traffic dust and coal combustion were also responsible for the origin of anomalous elements Cd, Cr, and Ni (0.004–1.8, 0.76–590, and 0.14–372 mg/kg, respectively). 90.15 % of the Leizhou Peninsula plants were not obviously contaminated, yet the comparison between the data collected in 1997 and 2018 allows us to speculate that Ni in the studied soils will reach the risk screening value in 7 years, followed by Cr and Cu in 39 and 92 years, respectively.

1. Introduction

China's 1:250,000 multi-target regional geochemical survey has covered most of the land area, such as urban, river, lake, grassland, shallow sea, and other ecosystems, since its implementation in 1999 (Xie et al., 1989, 1997; Darnley, 1995). An abundance of high-quality geochemical data has been obtained, providing robust support for further regional eco-geochemical evaluation (e.g., Yang et al., 2005). The survey not only identified the soil geochemical quality in China's major agricultural and industrial areas, but also pointed out several problems in some of the main agricultural production bases, such as the excessive usage of various organic fertilizers, abnormal enrichment of heavy metals in soils, accelerated soil acidification, and deterioration of soil environmental quality (e.g., Hu et al., 2017; Eli et al., 2020; Guo et al., 2021). To identify and solve these problems, regional eco-geochemical surveys have been conducted with the main research contents of heavy metal sources, migration, ecological effects, and ecological early warning (Yang et al., 2008; Chen et al., 2008; Xi, 2008; Zhang et al., 2018; Li et al., 2021a; Tian et al., 2022).

To date, although regional eco-geochemical surveys have been well carried out in most provinces in China, the Leizhou Peninsula is still in a gap in terms of land quality geochemical surveys and regional eco-geochemical surveys (Chai et al., 2019). Since the reform and opening-up policy, the Leizhou Peninsula has achieved rapid social and economic development, yet it has also paid a heavy price in terms of resources and the environment, e.g., the decline in the quantity and quality of natural resources has seriously affected food security production and the cleanliness of the human environment, and restricted the sustainable and rapid socio-economic development of the region (Li et al., 2014; Dou and Li, 2015; Cai et al., 2019; Wang et al., 2020a). To improve the ecological environment and provide a scientific basis for economic and social development and planning, we carried out a geochemical survey of land quality over the Leizhou Peninsula. The survey covered an area of 12,490 km^2, and systematically collected surface soil, deep soil, and mudflat sediment samples using a two-layer grid arrangement. Meanwhile, bulk agricultural crops, irrigation water,

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atmospheric sediment, and rock samples were also collected in conjunction with the characteristics of the study area. The results obtained in this study will largely fill the gap in land quality and eco-geochemical evaluation in the Leizhou Peninsula. Together with the data we have gained in 1997, this study could help us to make a valuable prediction to assess the spatial and temporal evolution trend of heavy metal elements contents in the Leizhou Peninsula.

2. Overview of the study area

The study area is located in the southernmost part of mainland China and at the intersection of Guangdong, Guangxi, and Hainan provinces. It is surrounded by the South China Sea to the east, the Qiongzhou Strait to the south, and the Beibu Gulf to the west (also named as "Tonkin Gulf") (Fig. 1). The geographical coordinates of the study area are 20°12′-21°55′N and 109°39′-110°57′E. The administrative area of the Leizhou Peninsula includes four districts (Chikan, Xiashan, Mazhang, and Potou), three county-level cities (Leizhou, Lianjiang, and Wuchuan), and two counties (Xuwen and Suixi). The terrain of the study area is dominated by plains and hills. Geotectonically, it belongs to the South China Fold System, next to the Yunkai Massif and the Yuezhong Massif of the Cathaysian Block (Zhang et al., 2018). Exposed stratigraphy in the Leizhou Peninsula comprises the Pre-Cambrian, Silurian, Devonian, Carboniferous, Jurassic, and Quaternary sequences, of which the Quaternary strata are the most widely distributed, accounting for 78 % of the total area (Guangdong BGMR, 1988). Effusive rock is mainly of the Pleistocene basalt and basaltic pyroclastic rock, which are mostly distributed in the southern part of the Leizhou Peninsula. Mesozoic intrusive rocks (e.g., granite) are locally distributed in the northwest of the study area (Guangdong BGMR, 1988).

There are six types of soils in the Leizhou Peninsula, including red soil, brick red soil, acidic sulphate soil, coastal salt soil, marsh soil, and rice soil, with red soil predominating. The distribution characteristics of these soils are: (1) brick red loam in the area south of 20°40′N, which accounts for more than half of the total land area and is the most important soil type; (2) red loam in the area north of 20°40′N; (3) rice

Fig. 1. The land usage map of the Leizhou Peninsula, Guangdong Province.

soils are widely distributed throughout the study area, with large areas from Lianjiang to Anpu Port, southwest of Wuchuan City, and around Leizhou City; (4) acidic sulphate soils and coastal saline soils in the coastal areas; (5) tidal sandy mud soils on the two sides of the Jiuzhou River and the Jianjiang River (Fig. 1). According to the data of the Second National Land Survey of Guangdong Province, the arable land in the Leizhou Peninsula covers an area of 4643.8 km^2, accounting for 35.07 % of the total land area, and the land usage is mainly arable land, forest land, and water (Guangdong PSCO, 1993; Fig. 1).

3. Sample collection and analytical testing

3.1. Soil samples

Surface and deep soil samples were collected using a two-layer grid method. Sampling points are designed to be evenly distributed in the study area under the premise of being far away from human activities and the thick overlay. The high-density sampling allows the collected samples to represent the main soil type within the sampling grid. The sampling density for surface soil samples in the terrestrial area was 0.25 to 2 points/km^2, with 2 points/km^2 for urban areas and 1 point/4 km^2 for mudflats. Sampling depths ranged from 0 to 20 cm for surface samples collected by box-type stainless steel grabs and anchor sampling grabs. Deep soil samples with a depth of 150 to 200 cm were collected continuously in the middle of the grid by the DDC-Z-2 vibratory sampler. Deep soil samples were collected with a density of 1 point/4 km^2 on land and 1 point/16 km^2 on mudflats. All the samples from the terrestrial area were combined into one analysis sample for every 4 km^2, while samples from the mudflats were analyzed singularly. The samples were naturally dried, sieved through 20 mesh (<0.84 mm) and sent to the laboratory to be processed to 200 mesh (<0.074 mm) for analysis. Soil pH was determined by passing the samples through a 10 mesh (<2 mm) sieve.

3.2. Other samples

In order to study the distribution characteristic, source, migration, and transformation of toxic and hazardous elements in the special agricultural area and element anomalous area of the Leizhou Peninsula, we also collected lots of rock, water, dry and wet deposition, and bulk crop samples. The sampling location was based on the geological background, soil types, land use patterns, content of heavy metal elements, and soil physical and chemical properties in the study area. All samples were collected and processed according to the Technical Requirements for Regional Eco-geochemical Evaluation (MLRC, 2015).

3.3. Sample analysis and quality control

The test methods and quality requirements for elemental indicators of conventional samples of rocks, soils, and water strictly follow the "Specification for Multi-Target Regional Geochemical Investigations", "Technical Requirements for Regional Ecogeochemical Evaluation", and "Technical Requirements for Analysis of Samples for Ecogeochemical Evaluation" of the China Geological Survey (MLRC, 2014, 2015).

Soil, rock, and atmospheric dry settlement samples were mainly analyzed using a combination of XRF, ICP-MS, and ICP-OES, and also combined with other advanced and sensitive special analytical instruments (Table 1). For plant samples, plasma spectrometry was used as the main method, supplemented by plasma mass spectrometry, atomic fluorescence spectrometry, ion-selective electrodes, volumetric methods, and gravimetric methods. For water and atmospheric wet settlement samples, plasma mass spectrometry and plasma spectrometry were used as the main methods, supplemented by atomic fluorescence spectrometry, spectrophotometry, volumetric methods, ion-selective electrode methods, catalytic colorimetric methods, turbidimetric methods, and potentiometric methods. Sample testing is carried out by the Guangdong Geological Experimental Testing Centre, and the quality of the samples is monitored using a combination of external laboratory and internal quality control methods. The detection limits of the analyzed elements are required to be close to or lower than the abundance of crustal elements, the accuracy (ΔlgC) is controlled at 0.10–0.12, the precision (RSD) is controlled at 10 %–20 % and the reporting rate is above 98 %.

Table 1
Package of test and analysis methods.

Analysis methods (10 ways)	Number of items	Determination of 54 elements
X-ray fluorescence spectrometry (XRF)	25	Na_2O, MgO, Al_2O_3, SiO_2, K_2O, CaO, TFe_2O_3, Ba, Br, Ce, Cl, Ga, Mn, Nb, P, Pb, S, Sc, Sr, Ti, V, Y, Zn, Zr, (Cr), (Co), (Cu), (La), (Ni), (Rb), (Th)
Plasma mass spectrometry (ICP-MS)	15	Au, Be, Bi, Cd, Co, La, Li, Mo, Rb, Sb, Th, Ti, U, W, (Ce), (Pb)
Plasma spectrometry (ICP-OES)	9	Cr, Cu, Ni, (Mn), (Pb), (Zn), Ba (<60 × 10^{-6}), (V), B (>200 × 10^{-6}), (P), (Ti), (Sr), (Na_2O), (K_2O)
Atomic fluorescence spectrometry (AFS)	4	As, Hg, Se, Ge
Emission Spectrometry (ES)	3	Ag, Sn, B
Colorimetric method (COL)	1	I
Ion Selective Electrode Method (ISE)	2	F, pH
High-frequency infrared carbon and sulfur analyzer infrared absorption method	2	TC, S (>800 × 10^{-6})
Volumetric method (VOL)	2	N, Corg.
Weight method	1	SiO_2 (>95 %)

Note: The item "()" is a supplementary certification scheme.

3.4. Soil environmental quality evaluation methods

According to the screening and control values of soil pollution risk on agricultural land, as specified in the "Soil Environmental Quality Soil Pollution Risk Control Standard (Trial)" (GB15618-2018) (MEEC, 2018), soil environmental quality was evaluated for soil environmental quality grading of heavy metal elements in the soil. Based on single-indicator soil environmental geochemical grading, the comprehensive soil environmental geochemical grade of each evaluation unit is equal to the worst grade of the environmental grade delineated by the single indicator. The classification standards of the quality evaluation are provided in Tables 2 and 3.

4. Results and discussion

4.1. Elemental geochemical characteristics

4.1.1. Elemental content characteristics of soil samples

Geochemical benchmark values refer to the natural range of elemental content in a medium in the circumstance of a subsurface environment, i.e., the content of elements without the influence of human activity (Tian et al., 2022), and thus in this paper refers to the content of elements in deep soils. Geochemical background value is the normal range of variation in elemental content within a certain area or statistical unit, reflecting the material composition characteristics of a particular geochemical evolution (Wang et al., 2020b; Li et al., 2021b; Yuan et al., 2021; Yoon et al., 2022). They are characterized in this paper by the statistical characteristic values of elemental content in surface soils. In general, the chemical composition of topsoil (0–20 cm), which has undergone a long period of soil formation and is influenced by human activity, inherits some of the characteristics of the parent material, but also differs significantly from it.

Regional geochemical benchmark values are regional differences in

Table 2
Classification standard of soil pollution risk screening value and control value of agricultural land.

Number	Elements	Risk screening value and risk control value				
			pH ≤ 5.5	5 < pH ≤ 6.5	6.5 < pH ≤ 7.5	pH > 7.5
1	Cd	Screening value	0.3	0.3	0.3	0.6
		control value	1.5	2.0	3.0	4.0
2	Hg	Screening value	0.5	0.5	0.6	1.0
		control value	2.0	2.5	4.0	6.0
3	As	Screening value	30	30	25	20
		control value	200	150	120	100
4	Pb	Screening value	70	90	120	170
		control value	400	500	700	1000
5	Cr	Screening value	150	150	200	250
		control value	800	850	1000	1300
6	Cu	Screening value	50	50	100	100
		control value	–	–	–	–
7	Ni	Screening value	60	70	100	190
		control value	–	–	–	–
8	Zn	Screening value	200	200	250	300
		control value	–	–	–	–

Unit: mg/kg.

Table 3
Classification table of soil environmental geochemical grade.

Soil environmental geochemical grade	First class	Second class	Third class
contamination risk	pollution-free risk soil	risk controllable soil	high-risk soil
Division method	Ci ≤ *Si*	*Si* ≤ *Ci* ≤ G*i*	Ci > G*i*

Note: Ci is the measured concentration of the i index in the soil, Si is the risk screening value, and Gi is the risk control value.

elemental content determined by the material composition of the soil. This difference is often determined by soil-forming parent material properties and natural landscape conditions (Li et al., 2021b; Xin et al., 2021; Hussain and Hoque, 2022). In this paper, this regional difference can be seen by comparing the results of deep soil samples with crustal elemental abundances (Fig. 2a). The results show that the elements depleted in the deep soils of the study area are mainly Fe-group elements, pro-Cu elements, and rock-forming mineral elements, including K_2O, MgO, Na_2O, CaO, Cr, Ni, Co, TFe_2O_3, Mn, V, La, Sc, Rb, Sb, Cu, Au, Ag, Zn, Hg, Sr, Ba, Ti, Cd, S, F, P, and Cl. The enriched elements are Zr, Th, Bi, W, Sn, As, Pb, Se, N, B, I, etc. (Fig. 2a). Among them, the enrichment coefficients of N, I, and Se are as high as 15.2, 5.2, and 4.3, respectively. The elements that are closer to the crustal elemental abundance are SiO_2, Al_2O_3, Nb, Be, Li, Y, Mo, Ge, Ga, and TC (Fig. 2a). Likewise, compared to the crustal element abundance (52 indicators), the surface soils in the study area are enriched in Bi, N, TC, Se, I, B, Zr, Sn, Th, As, W, Pb, depleted in P, K_2O, Na_2O, CaO, Al_2O_3, MgO, TFe_2O_3, Mn, Co, Ni, Ti, V, Cr, Cu, Zn, Hg, Au, Ag, Sb, Mo, Be, Cd, Li, Ti, Ga, La, Sc, Ba, Cl, Rb, Sr, and in comparable amounts of SiO_2, Ce, Y, U, Br, Nb, Ge, S (Table 4). The enriched elements are mainly beneficial nutrients, with enrichment factors >2 for Corg, Bi, N, TC, Se, I, B, and Zr. The depleted elements are mainly rock-forming minerals that are easily lost. The enrichment factor is <0.5 for Sc, Ti, V, Cr, Zn, TFe_2O_3, Cd, Ba, Cl, Au, Cu, Rb, K_2O, Ni, Co, Mn, MgO, Sr, Na_2O, and CaO. The geochemical parameters of the surface and deep soil samples in the study area are shown in Table 4 and Fig. 2b. Geochemical parameters of the surface sediment samples from the coastal mudflat area are shown in Table 5. Furthermore, the mudflat soils are alkaline with a mean pH of 7.9. Most of the elemental background values are lower than the region-wide background values, enriched in Cl, Na_2O, Co, Mn, Br, K_2O, MgO, Sr, CaO, Rb, Ba, As, and depleted in Zn, Pb, TFe_2O_3, La, Y, U, Ce, Cr, Au, V Sb, Sc, Ag, Th, Nb, Cd, Cu, Zr, Sn, Ti, W, Mo, Ga, Bi, Al_2O_3, P, Se, N, TC, Hg, Corg. The enriched elements are mainly halogen group elements and alkali metal elements. The depleted elements are dominated by iron group elements and beneficial nutrients.

Generally, the surface soils in the study area are enriched in P, Corg, N, TC, S, Br, Cd, Hg, CaO, and Cl, and depleted in Ge, Sr, MgO, Al_2O_3, Mo, I, Ga, Cu, Sc, V, TFe_2O_3, Be, Ni, Cr, K_2O, Rb, and Co when compared with the regional soil benchmark values. This enrichment is evident for pro-biotic and environmental elements, with enrichment factors >3 for P, Corg, N, and TC. On the contrary, the depletion is evident for Fe-group elements and rock-forming minerals, and the most depleted element is Co, with an enrichment factor of 0.4. The variability in elemental enrichment between surface and deep-layer soils (e.g., N, I, and Se) is generally due to geological, geochemical, and soil-forming interactions. The surface soils enriched in P, N, Corg, and TC are mainly caused by bioconcentration and the influence of human activities, while the depletion in elements such as TFe_2O_3, Al_2O_3, and K_2O in the surface and deep layer soils is mainly caused by leaching.

4.1.2. *Elemental geochemical distribution characteristics*

The Leizhou Peninsula is a typical geochemical landscape area with significant variability in the spatial distribution of elements (Fig. 3). In terms of the geochemical distribution characteristics of the elements, the high elemental combinations Ag, As, Au, B, Bi, Ce, Cd, K_2O, Pb, Rb, Sb, Sn, Th, Ti, U, W, Zr, etc. are mainly distributed in the northern part of the study area. By contrast, the elemental anomalies in the southern part are P, SiO_2, Zr, Fe_2O_3, V, Cr, Cu, Mn, Ni, Zn, Co, Ti, Al_2O_3, Sc, Nb, I, etc. Moreover, some elements of the Fe group and environmentally related elements show moderately weak or moderately strong anomalous bands throughout the region. Generally, according to the distribution characteristics of the regional elements, the Leizhou Peninsula can be divided into two domains: W, Sn, Bi, Th, U, La, Y, Ce, Au, Ag, Pb, Sb, K_2O, Ti rich zone in the north (I) and Fe_2O_3, V, Cr, Cu, Mn, Ni, Zn, Co, Ti, Al_2O_3, Sc, Nb, I rich zone in the south (II) (Fig. 3). These two zones can be further subdivided into 7 subzones based on the further refinement of the characteristic elemental assemblages, and the similarity of the characteristic elemental assemblages, soil-forming parent materials, or soil-forming environments in the study area. Detailed element combinations of the different subzones are shown in Fig. 3.

4.2. *Eco-geochemical evaluation*

4.2.1. *Characteristics of heavy metal concentrations in surface soil*

It can be seen from Table 6 that except for some individual anomalies in local areas (Fig. 4a), the content of heavy metals in the surface soil of the Leizhou Peninsula is generally low. For example, the content of Cd, Hg, and Zn in the surface soil of the Leizhou Peninsula is 0.074 mg/kg, 0.070 mg/kg, and 51.998 mg/kg, respectively, lower than those in other regions or countries (Table 6). The average concentration of Pb in the surface soil of Leizhou Peninsula is lower relative to that in the Pearl River estuary sediments and the England and Wales agricultural soils, but close to the Pb content in the shipbreaking area of Sitakund Upazilla sediments (Table 6). The average concentration of Cu is 29.467 mg/kg, which is similar to the Cu content in Danube Delta sediments and lower

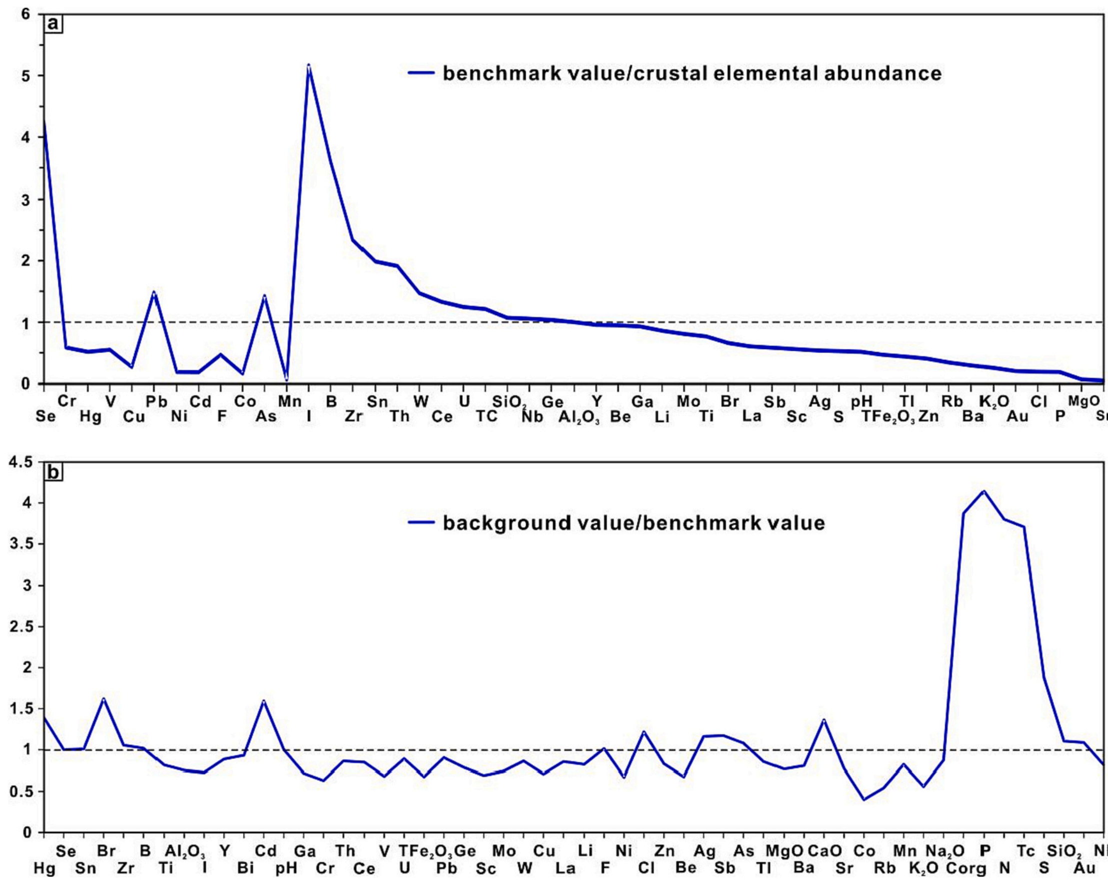

Fig. 2. (a) Crustal elemental abundance (Taylor and McLennan, 1985; Rudnick and Fountain, 1995) normalized for the Leizhou Peninsula soil element benchmark value; (b) benchmark value normalized for the Leizhou Peninsula soil element background value.

Table 4
Statistics of soil geochemical benchmark and background values from the Leizhou Peninsula.

Element (indicator)	Number of samples	Background values	Base value	Element (indicator)	Number of samples	Background values	Base value
Ag	808	0.050	0.043	Pb	815	16.1	17.8
As	836	3.38	3.13	Rb	790	14.5	26.7
Au	842	0.87	0.80	S	623	394	210
B	854	47.7	46.8	Sb	844	0.41	0.35
Ba	852	93	115	Sc	870	7.0	10.1
Be	860	0.83	1.23	Se	834	0.34	0.34
Bi	835	0.28	0.30	Sn	850	3.41	3.38
Br	807	4.7	2.9	Sr	784	17.6	22.7
Cd	833	0.059	0.037	Th	833	9.6	11.1
Ce	837	48.6	57.3	Ti	864	0.400	0.490
Cl	710	65.3	53.6	Tl	840	0.18	0.21
Co	838	1.6	4.0	U	807	1.89	2.12
Cr	869	40.7	64.5	V	870	52.3	76.74
Cu	871	12.0	16.9	W	826	1.40	1.62
F	824	213	210	Y	854	20.3	22.9
Ga	872	12.0	16.7	Zn	870	31.7	38.2
Ge	867	1.14	1.45	Zr	854	321	304
Hg	843	0.064	0.046	SiO_2	872	73.30	66.38
I	776	2.25	3.10	Al_2O_3	872	11.78	15.65
La	820	20.11	23.50	TFe_2O_3	870	2.63	3.89
Li	786	14.8	18.0	MgO	790	0.23	0.30
Mn	721	65	79	CaO	725	0.15	0.11
Mo	846	0.78	1.05	Na_2O	725	0.07	0.08
N	848	1041	274	K_2O	846	0.29	0.52
Nb	856	16.3	20.0	TC	787	1.26	0.34
Ni	871	11.20	16.70	Corg	786	1.20	0.31
P	797	936	226	pH	736	5.15	5.14

Units: Au in ng/g, Oxide, TC, Corg in %, pH dimensionless, rest in μg/g. Number of samples is the number of samples after exclusion.

Table 5
Summary of geochemical background values for coastal mudflat surface sediments.

Element	Mudflats (215)	Element	Mudflats (215)
Ag	0.031	Pb	12.5
As	4.19	Rb	22.1
Au	0.57	S	445
B	49.1	Sb	0.26
Ba	121	Sc	4.4
Be	0.78	Se	0.10
Bi	0.11	Sn	1.90
Br	11.8	Sr	29.8
Cd	0.034	Th	5.8
Ce	32.6	Ti	0.220
Cl	2445.0	Tl	0.15
Co	5.4	U	1.29
Cr	27.3	V	34.14
Cu	6.8	W	0.72
F	184	Y	14.0
Ga	5.6	Zn	25.1
Ge	1.06	Zr	180
Hg	0.014	SiO_2	85.30
I	2.01	Al_2O_3	4.29
La	15.50	TFe_2O_3	2.03
Li	12.0	MgO	0.51
Mn	196	CaO	0.24
Mo	0.38	Na_2O	0.50
N	293	K_2O	0.69
Nb	9.5	TC	0.35
Ni	12.21	Corg	0.24
P	314	pH	7.92

Note: Units are μg/g, Au is ng/g, Oxide, TC, Corg are %, pH is dimensionless.

than that in other regions. Other heavy metal contents in the surface soil of the Leizhou Peninsula and their comparison with those in other regions or countries are also presented in Table 6. These comparisons can give a more objective understanding that the soil environment of the Leizhou Peninsula is generally good.

4.2.2. Soil environmental quality assessment

The evaluation results are shown in Fig. 4. In general, the overall soil environmental quality in the study area is good with ~9168.07 km^2 of pollution-free risk soil that was concentrated in the northern part of the Leizhou Peninsula. The area of risk-controllable soil is 3318.18 km^2 and is mainly distributed in the southwest of Suixi County, southern Leizhou City, and most areas of Xuwen County (Fig. 4). The distribution range of the risk-controllable soil highly coincides with the basalt outcrop area (Fig. 4; Guangdong BGMR, 1988). High-risk soil is only distributed in a single point in the northern part of Lianjiang City, which may be affected by nearby mining activity.

4.2.3. Natural and artificial sources of abnormal heavy metal elements

Soils are derived from the weathering products of rocks, and therefore the study of the relationship between soils and their parent rocks is conducive to determine the source of heavy metal contaminants (Liu et al., 2022). The distribution of geological units in the Leizhou Peninsula is relatively clear, with Paleozoic-Mesozoic granites, clastic rocks (e.g., Devonian Xindu shales), and low-grade metamorphic rocks (e.g., Yunkai Group) in the north; Holocene alluvium together with some basalt outcrops (mostly in the Huguangyan Group; Fig. 4b) and Pleistocene fluvial-facies sedimentary rocks (e.g., Zhanjiang Group) in the central; and mostly Shimaoling basaltic volcanic rocks in the south (Guangdong BGMP, 1988; Fig. 4b). Based on the distribution characteristics of the elements in typical strata and rocks, we found that the content of heavy metal elements varies highly in different soil-forming parent rocks (Table 7). For example, Cr, Ni, and Cu are abundant in basaltic rocks, while As, Pb, and Cd, as well as Pb and Zn, are rich in the Devonian Xindu shale, Late Jurassic diorite granites, and Early Ordovician diorite granites, respectively (Table 7). These felsic and clastic rocks are mainly distributed in the northern Leizhou Peninsula as mentioned above (Fig. 4b). Compared with the abundance of crustal elements (Li, 1976), the As content in the Xindu shale is 19.8 times higher, suggestive of a strong enrichment (Table 7). The Cr, Ni, and Cu elements in basalts have the highest enrichment coefficient of 2.7 relative to the crustal elements (Table 7). More importantly, by comparing the background values of elements in the collected soil samples with the mean values of elemental contents in the adjacent soil-forming parent rocks, we found that, except for As and Hg, these two values have a good co-elimination relationship (Fig. 5). This indicates that the content of Cr, Ni, Cu, and Cd in the study area is closely related to the geological background and is controlled by the soil-forming parent material to some extent.

In addition, the heavy metal pollutants in the soil environment that are carried by high-intensity human activities cannot be ignored (Hasan et al., 2013a, 2013b). In this study, we have calculated the input fluxes of exogenous pollution, such as dry and wet atmospheric deposition and irrigation (Tables 8 and 9), following the calculation formula below:

$$F = Qt/S = (Qs + Qi)/S = (V^{*}Cs + M^{*}Ci)/S,$$

F is the annual sedimentation flux (mg. M^{-2}. a^{-1}), Qt is the total annual sedimentation (mg/a), S is the sampling area (m^2), Qs is the annual sedimentation of the wet sample (mg/a), Qi is the annual sedimentation of the dry sample (mg/a), V is the total volume of the solution (L/a), Cs is the element concentration of the solution sample (mg/L), M is the total annual sedimentation (g/a), and Ci is the element mass fraction of the dry sample (mg/g).

Dry and wet atmospheric settling pollutants in the Leizhou Peninsula were mainly Zn, Cr, and Ni, and the arrangement order of element settlement flux density was Zn > Cr > Ni > As > Pb > Hg > Cd (Table 8). Regionally, atmospheric deposition in the Wuchuan-Wuyang area is relatively enriched in Zn, Ni, and As, while it is relatively enriched in Hg and Pb in the Suixi-Chengyue area (Fig. 1). The sedimentation flux density of Hg and Pb in the Suixi-Chengyue area is 2.6 and 1.5 times higher than that in the Wuchuan-Wuyang area, respectively (Table 5). Factor analysis of heavy metal elements of atmospheric dry and wet settlement materials shows that two factors (F1 and F2) can be selected as the main component factors in the whole region (Table 9). F1 is composed of Cd, Cr, Cu, Pb, Ni, Hg, and [As], and F2 comprises As and [Cr, Pb, Ni], where [ ] indicates negatively correlated (Table 9). According to the results of the agricultural geological survey on the Pearl River Delta of China (PRD) (Lai, 2005), endogenous dust can be divided into several types. The endogenous dust in the PRD are as follows: ① road traffic dust (DJ): Cu, Zn, Pb, Cd, Hg, Cr; ② metallurgical ceramic dust (YT): Cd, Pb, F, S; ③ coal combustion dust (RM): Ni, S, Cd, As, Pb, Cr; ④ chemical industry dust (HG): Zn, S, Pb, Cd; ⑤ construction dust (JZ): CaO, Pb, Cd; ⑥ tail gas dust (WQ/WC): Pb, Cr. According to the division of these endmembers of the endogenous dust in the PRD, F1 should be most likely the road traffic dust, while F2 may be the coal combustion dust. Thus, we can conclude that the heavy metal elements imported into the soil by dry and wet atmospheric deposition mainly originated from road traffic and coal combustion.

In addition, as presented in Table 10, the quality of irrigation water in the study area seems good, especially in the Wuyang area where the content of heavy metal elements is below the detection limit. Therefore, irrigation water should have a mild effect on the elemental content of the surface soil.

4.2.4. Evaluation of the ecological effects of plants

Previous studies have shown that heavy metals can migrate into agricultural products causing harm to human health (Hasan et al., 2019). A total of 132 plant samples were collected from the study area and tested for 10 indicators including As, Cd, Hg, Pb, Cr, F, Cu, Zn, Ni, and Se. A total of 13 plant samples were found to exceed the limits according to the National Food Safety Standard for Contaminants in Food

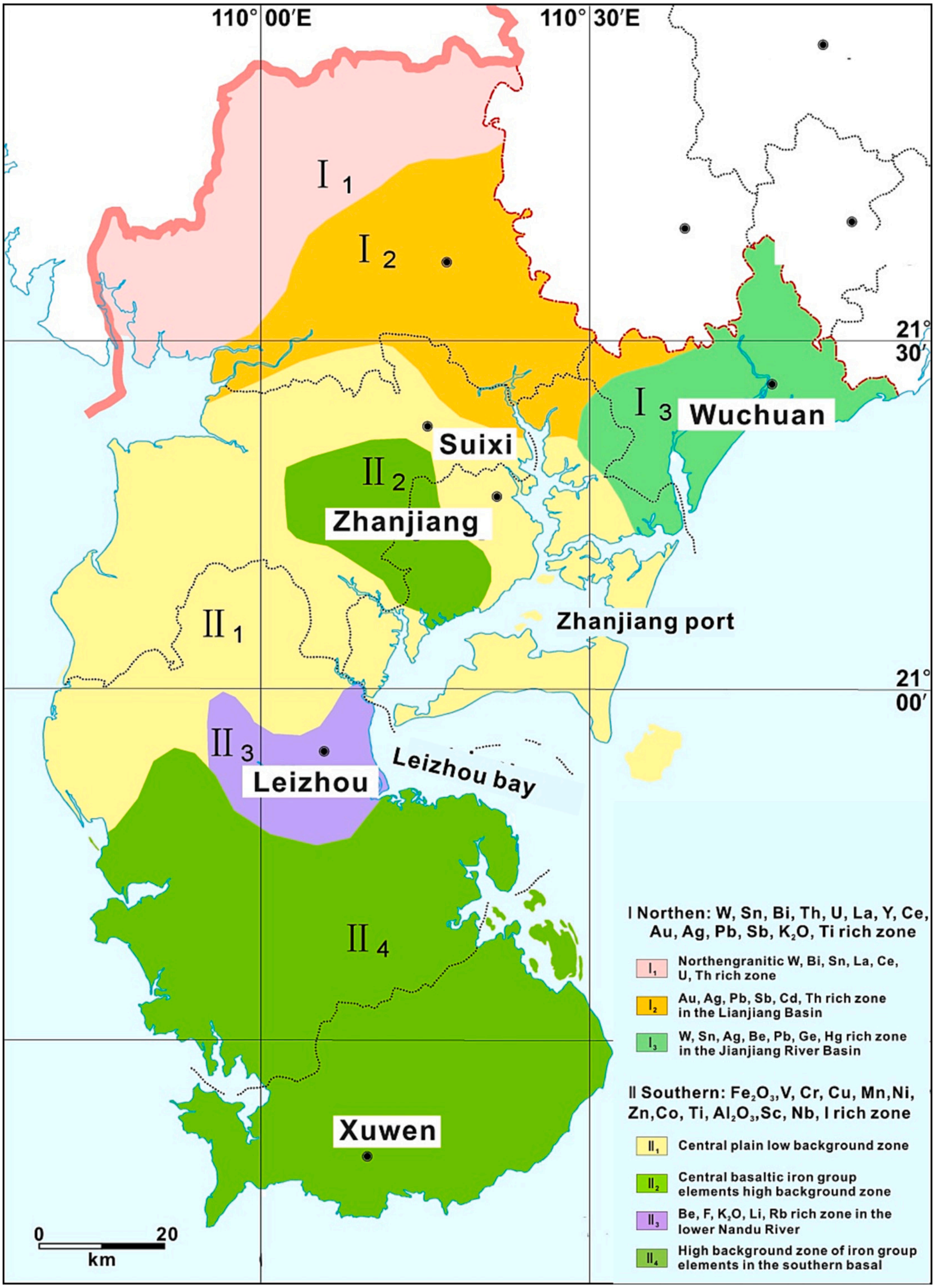

Fig. 3. Geochemical zoning map of the Leizhou Peninsula.

(GB2762-2017), with an exceedance rate of 9.85 %. Ten of these contaminated plants are rice, one is red river orange, and two are peanuts, with excessive elements of Cr, Pb, and Cd (Table 12). The remaining elements in these plant samples have a normal content (Tables 11 and 12).

As can be seen from Fig. 6, the place with the highest rate of excessive plant pollution is the Xuwen City, with Pb and Cr heavy metal elements exceeding the limit standards The number of exceedance cases for rice in the Xuwen City with excessive Pb and Cr levels was 7 and 3, respectively. Moreover, there are also two cases for peanut samples, which have higher Cr contents than the limit standards. By contrast, the Suixi-Chengyue region has the best quality samples, with no samples exceeding the limits for contaminants in food (Fig. 6).

4.2.5. Prediction of spatial and temporal trends in the environmental quality of soil heavy metals

In 1997, the project "1:200,000 sediment measurement of Zhanjiang water system" was carried out in the Leizhou Peninsula. In addition to the water-system samples, a large number of soil samples were collected in the southern area, which could provide an important reference for

Table 6
Heavy metal contents in the study area and comparison with the results presented in the published literature (unit in mg/kg).

Region	Parameter	Cd	Hg	As	Pb	Cr	Cu	Ni	Zn
Leizhou Peninsula (this study)	Mean	0.074	0.070	4.037	19.576	108.472	29.467	56.007	51.998
	Range	0.004–1.8	0.002–1.09	0.32–108	2.8–323	0.76–590	0.13–354	0.14–372	3.1–257
Pearl River Estuary (Pan and Wang, 2012)	Mean	5.6	0.33	33.1	105.9	118.1	81	–	140
Sara Union, Export Processing Zone Area, Ishwardi, Pabna (Islam et al., 2022)	Mean	–	–	–	–	3692.39	110.48	354.16	236.08
Ship breaking area of Sitakund Upazilla, Chittagong (Hasan et al., 2013a)	Mean	0.24	–	6.81	18.09	658.45	189.18	32.64	355
Danube Delta (Marindrescu et al., 2022)	Mean	0.23	–	–	27.4	101.7	28.7	54.6	76.8
England and Wales (Nicholson et al., 2003)	Mean	1.9	1.0	3.1	54	7.5	57	16	221

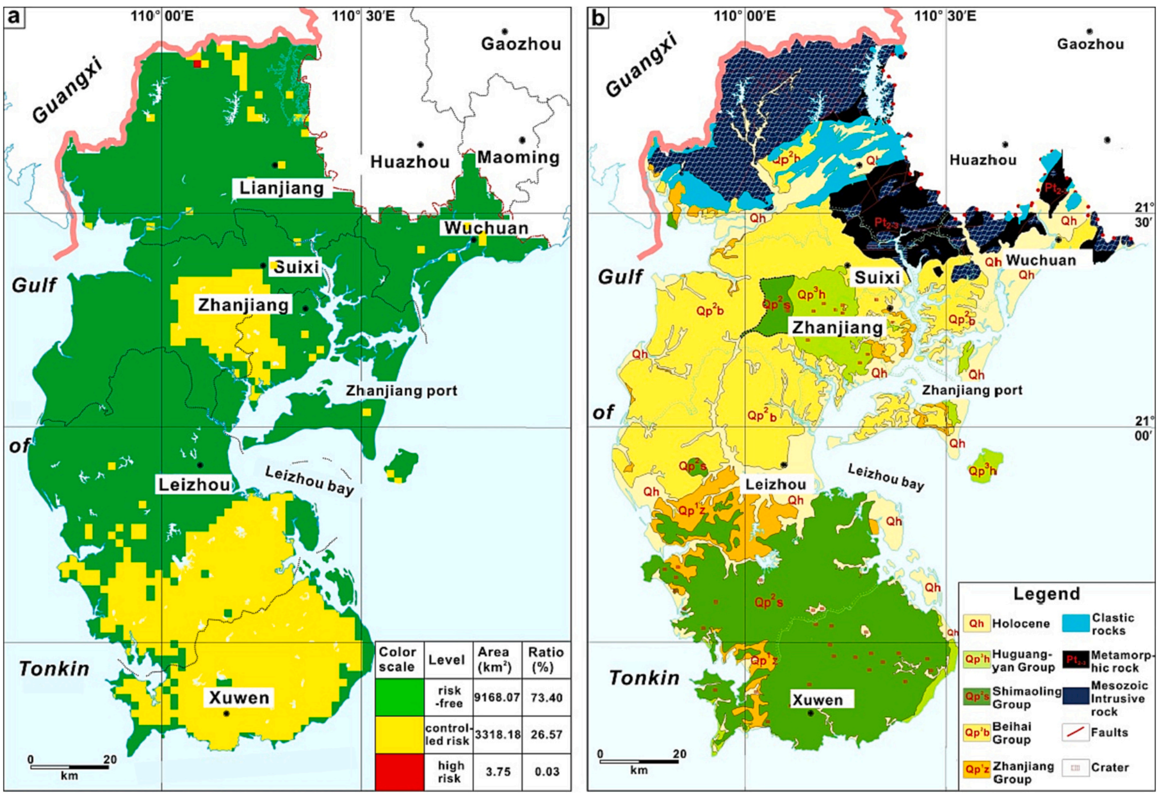

Fig. 4. (a) Comprehensive geochemical grading of the soil environment in the study area (this study); (b) Geological map of the Leizhou Peninsula, showing the exposure of sedimentary and magmatic outcrops (Guangdong BGMR, 1988).

Table 7
The heavy metal element content of rocks in the Leizhou Peninsula (unit: μg/g). The location of granite, metamorphic rocks, and shale are mostly distributed in the northern Leizhou Peninsula, as shown in Fig. 4b.

Stratigraphic lithology	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Late Jurassic diagenetic granite	0.32	0.52	7.96	5.10	0.0019	3.36	46.4	77.2
Early Ordovician diorite	0.55	0.12	4.74	4.17	0.0027	2.58	155.95	127.25
Basaltic rocks in the Shimaoling Formation	0.27	0.07	246.50	69.50	0.0036	206	2.78	123
Basaltic rocks in the Huguangyan Formation	0.32	0.09	303	63.10	0.0016	196	2.41	104
Devonian Xindu Formation shale	43.55	0.02	108.15	25.10	0.025	19.90	27.45	77.55
Yunkai Group metamorphic rocks	0.88	0.02	3.45	2.13	0.0075	0.74	25.8	10.80
Crustal elemental abundances (Li, 1976)	2.20	0.20	110	63	0.08	89	10	94

this study on the evolutionary trend of heavy metal content in the soils. The median of the content ratio of the two sets of samples was used as the evaluation index to evaluate the changes in the content of heavy metals (e.g., Cd, Cu, Hg, Pb, and Zn). The result is shown in Table 13.

The results show that over the past 21 years, the soil Cr, Ni, and Cu contents in the study area have increased, the Pb and As contents have decreased, and the Zn, Cd, and Hg contents have remained basically unchanged (Table 13). Especially, the average contents of Cr, Ni, and Cu in the water-system samples were 90.3, 49.6, and 25.7 μg/g in 1997, but then increased to 111.1, 57.5, and 30.2 μg/g in 2018.

It is also noted that the content of Cr, Ni, and Cu increased significantly in the area where basalt and basaltic rocks mostly outcropped, with an addition of 85.2 μg/g for Cr, 30.4 μg/g for Ni, and 13.5 μg/g for Cu. Consequently, the high content of Cr, Ni, and Cu elements in the Leizhou Peninsula soils should be partly controlled by the local basalt and basaltic rocks. It is consistent with that the basaltic rocks usually

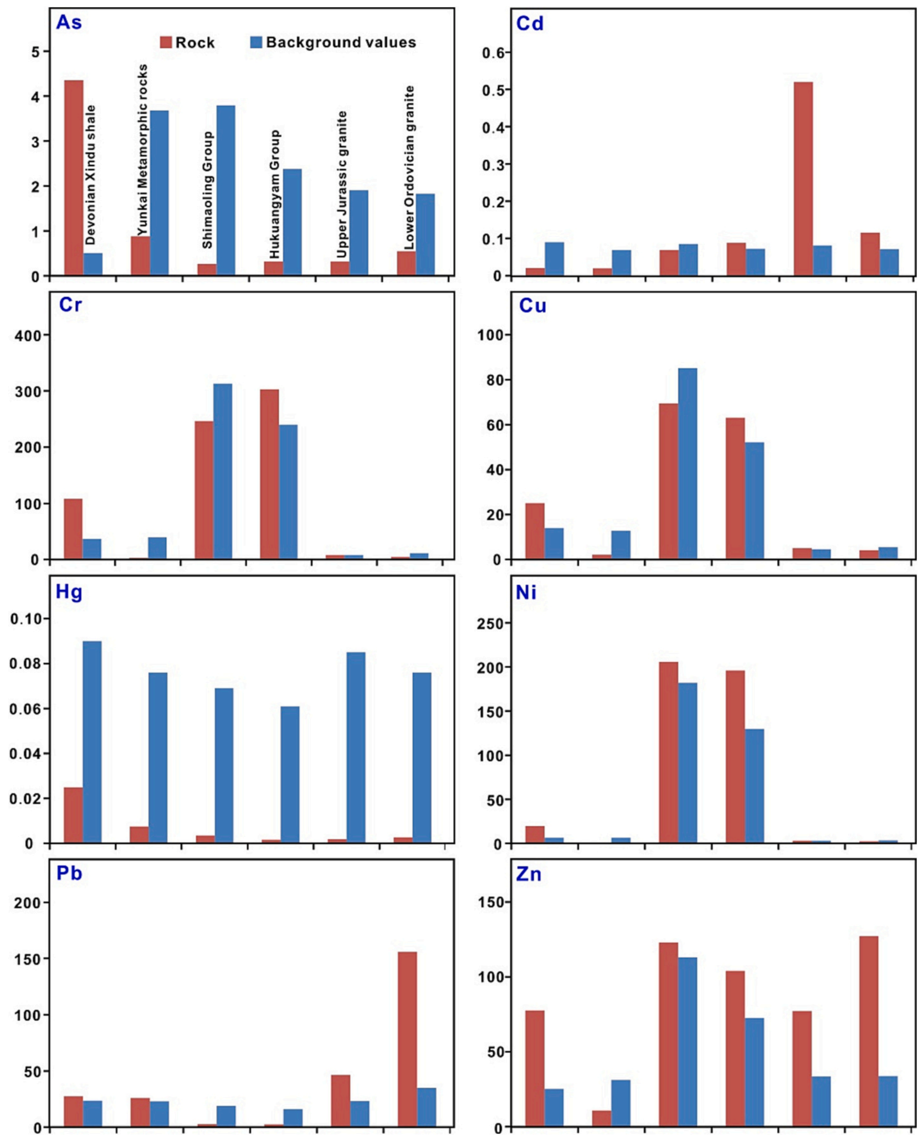

Fig. 5. Relationship between the heavy metal content of soil and soil-forming parent rock distributed in the Leizhou Peninsula. The data is shown in Table 4 and the location of each geological unit can be seen in Fig. 4b.

Table 8
Annual sedimentation flux density statistics for dry and wet atmospheric deposition in the Leizhou Peninsula.

Region	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Wu Chuan-Wu Yang	0.43	0.013	23.63	0.02	0.017	10.43	0.034	135.61
Suixi –Chengyue	0.17	0.016	32.70	0.01	0.044	2.15	0.052	20.93
Whole area	0.31	0.014	26.60	0.02	0.024	5.87	0.040	68.74

Unit: mg/(m^2 *a).

Table 9
Loadings of heavy metal factors in dry and wet atmospheric deposition of Leizhou Peninsula.

Element	F1	F2
As	−0.242	0.876
Cd	0.970	0.38
Cr	0.939	−0.283
Cu	0.859	0.219
Pb	0.966	−0.029
Zn	0.442	0.440
Ni	0.914	−0.279
Hg	0.838	0.409

have high contents of Cr, Ni, and Cu elements as shown in Table 7. In addition, for soils with a different type of use, Cr, Ni, and Cu increased significantly in garden soils by 55.6, 19.7, and 11.7 μg/g, followed by arable land (which includes both paddy and dry land) by 24.2, 10.4, and 5.1 μg/g respectively. The increase of heavy metals is not obvious in the urban land and forest land soils. Because agricultural development is dominant in the Leizhou Peninsula, it should therefore have some influence on the change in land quality.

As discussed above, the high background values of Cr, Ni, and Cu in the soils of the study area and the areas with large elemental variations are closely related to the distribution of the basaltic parent material. Assuming constant external conditions and elemental transport trans-

Table 10
Statistics of heavy metal elements in irrigation water in the Leizhou Peninsula.

Heavy Metal Element / Sample Number	Cr^{6+} ($\times 10^{-3}$)	As ($\times 10^{-4}$)	Hg ($\times 10^{-5}$)	Cu ($\times 10^{-3}$)	Zn ($\times 10^{-4}$)	Cd ($\times 10^{-4}$)	Pb ($\times 10^{-4}$)
WY001	<4	4	<5	2	32	<1	30
WY002	<4	5	<5	1.9	37	<1	28
WY003	<4	5	<5	2	42	<1	32
WY004	<4	5	<5	1.7	37	<1	26
WY005	<4	5	<5	1.8	36	<1	24
WY006	<4	3	<5	1.7	38	<1	27
WY007	<4	4	<5	2.3	31	<1	22
WY008	<4	6	<5	1.9	36	<1	24
CY01W	<4	<1	<5	1.3	<1	<1	2
CY02W	<4	<1	<5	0.6	<1	<1	1
CY03W	<4	<1	<5	0.8	<1	<1	1
CY04W	<4	<1	<5	2.1	<1	<1	6
XW01W	<4	<1	<5	1	<1	<1	<1
XW02W	<4	1	<5	1	<1	<1	<1
XW03W	<4	<1	<5	1	<1	<1	<1
XW04W	<4	<1	<5	<0.1	<1	<1	<1

Unit: mg /L.

Table 11
Contaminant limits in food.

Heavy metal / Food category	Lead	Cadmium	Mercury	Inorganic arsenic	Chromium
Cereals (brown rice)	0.2	0.1	0.02	0.2	1.0
Red River Orange (Fruit)	0.1	0.05	–	–	–
Peanuts (nuts)	0.2	0.5	–	–	1.0[a]

[a] Reference to bean standards.

formation processes in the Leizhou Peninsula, we proposed a computational model to estimate the time required for the average elemental content to reach the screening value for the risk of contamination of agricultural land with the following equation:

$$Ni = \frac{C1 - C2}{C2 - C3} * N$$

N_i: is the time for the element to reach the risk screening value, in years.
C_1: is the risk screening value for the element, using the tightest value for risk screening without regard to pH.
C_2: is the mean soil element content.
C_3: is the average of the elemental content of the water system.
N: is the time difference between the two tests, in this case, 21 years.

According to model calculations, the content of Ni in the study area is expected to reach the risk screening value after 7 years, followed by Cr and Cu after 39 and 92 years, respectively. It should be noted that the sampling media of the two projects are different, and the elemental input and output fluxes are unknown, so the conclusion drawn from this model is only for reference and does not represent the changes in heavy metal elemental content in the survey area. However, our model can indeed reflect that the southern Leizhou Peninsula is a high background area of heavy metal elements, and the monitoring of toxic and harmful heavy metal elements should be strengthened to grasp the dynamic changes of elements, to provide a basis for soil pollution control, land management, and soil remediation.

5. Conclusion

Our results show that the surface soils in the Leizhou Peninsula are characterized by enrichment of Bi, N, TC, Se, I, B, Zr, Sn, Th, As, W, and Pb, and depletion of P, K_2O, Na_2O, CaO, Al_2O_3, MgO, TFe_2O_3, Mn, Co, Ni, Ti, V, Cr, Cu, Zn, Hg, Au, Ag, Sb, Mo, Be, Cd, Li, Tl, Ga La, Sc, Ba, Cl, Rb, Sr, etc. The regional geochemical distribution of elements is restricted by the geological environment and anthropogenic economic activities, and shows different geochemical zoning: the northern part of the Leizhou Peninsula is enriched in W, Sn, Bi, Th, U, La, Y, Ce, Au, Ag, Pb, Sb, K_2O, Ti, etc., while the southern part is enriched in Fe_2O_3, V, Cr, Cu, Mn, Ni, Zn, Co, Ti, Al_2O_3, Sc, Nb, I, and other elements.

The results of heavy metal environmental quality show that the soil in the study area is mainly pollution-free risk soil (71.24 % in proportion), which is concentrated in the northern part of the Leizhou Peninsula. The risk-controllable soil accounts for 28.73 % of the total area and is mainly distributed in the southern Leizhou Peninsula, highly consistent with the basalt outcrop area. The high-risk soil is only distributed in the north of Lianjiang city in a single point, which is affected by nearby mines. The elemental result of the sedimentary and magmatic rocks in the study area and their relationship with that the soils shows that the

Table 12
Exceedance statistics for each type of plant sample.

Type		*inorganic arsenic		Chromium		Cadmium		Lead		Mercury		General	
		Pieces	Proportion	Pieces	Proportion	Pieces	Proportion	Pieces	Proportion	Pieces	Proportion	Pieces	Proportion
Hongjiang Orange	Qualified							20	95.24 %			20	95.24 %
	Exceeding standards	/	/	/	/	/	/	1	4.76 %	/	/	1	4.76 %
Peanuts	Qualified							6	85.71 %			5	71.43 %
	Exceeding standards	/	/	/	/	/	/	1	14.29 %	/	/	2	28.57 %
Rice	Qualified	/	/	103	99.04 %	101	97.12 %	96	92.31 %	/	/	94	90.38 %
	Exceeding standards	/	/	1	0.96 %	3	2.88 %	8	7.69 %	/	/	10	9.62 %

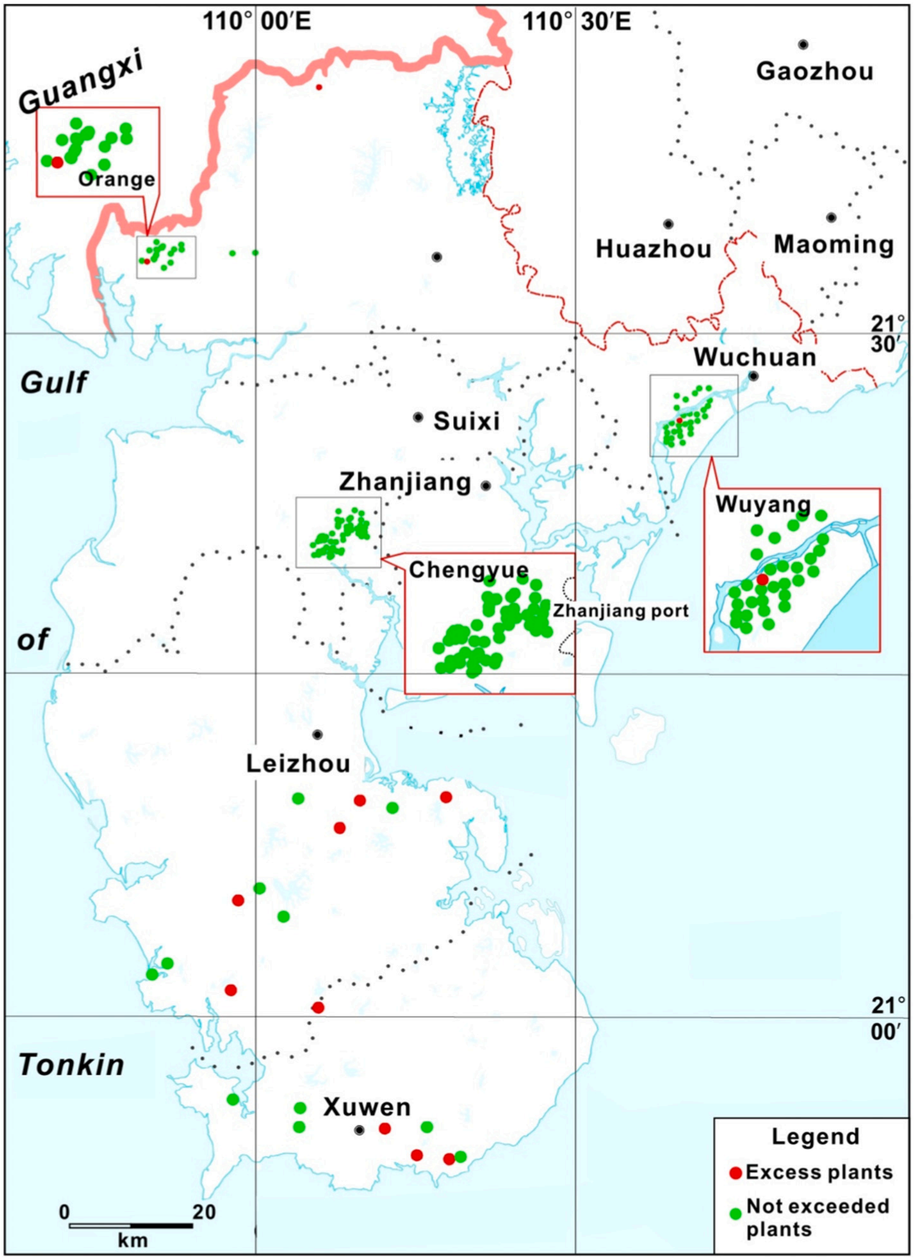

Fig. 6. Distribution of plant samples exceeding the standard in the Leizhou Peninsula.

Table 13
Estimated changes in soil heavy metal content in the Leizhou Peninsula. ** represents the quoted standard value, see Table 2 for details.

Element	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
1997 Water-system element content (μg/g)	4.22	0.07	90.29	25.72	0.071	49.62	28.28	49.27
2018 Soil element content (μg/g)	3.92	0.08	111.11	30.24	0.072	57.51	19.56	52.69
Soil risk screening value (μg/g)**	20	0.3	150	50	0.5	60	70	200
Soil element content up to Time to filter values (N_i[1])	*	839	39	92	8988	7	*	906

* Elemental content is reduced and not calculated.
1 N_i: is the time for the element to reach the risk screening value, in years.

natural rocks are the primary contributor of heavy metals to the soils. Moreover, atmospheric dust caused by high-intensity human activity was also responsible for the pollution of the soils in the Leizhou Peninsula.

In comparison with the data obtained in 1997, we found that Cr, Ni, and Cu contents of the soils in the study area had increased, Pb and As contents had decreased, and Zn, Cd, and Hg contents had remained unchanged. Under the prerequisite of the external conditions and elemental transport transformation processes for the Leizhou Peninsula soils remaining unchanged, Ni, Cr, and Cu are expected to reach the risk screening value after 7, 39, and 92 years, respectively. This long-term change allows us to calculate the increasing trend of the Cr, Ni, and

Cu contents in the Leizhou Peninsula soils. Our study not only gives a comprehensive eco-geochemical assessment on the Leizhou Peninsula, but also provides a good reference that highlights the importance of constant research in the same area.

CRediT authorship contribution statement

Tingting Li: Investigation, Conceptualization, Formal analysis, Writing – original draft, Writing – review & editing, Visualization
Xinchang Zhang: Writing – review & editing, Data interpretation, Conceptualization
Lili Jia: Investigation, Visualization
Xin Zhu: Investigation, Data interpretation
Min Xu: Writing – review & editing

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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