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Epigenetic geochemical dynamics and driving mechanisms of chemical elemental distribution patterns in soil in Southwest China*

© Hangxin Cheng^a, Min Peng^b, Chuandong Zhao^c, Wei Han^d, Huiyan Wang^e,
Qiaolin Wang^f, Fan Yang^g, Fugui Zhang^h, Chengwen Wangⁱ, Fei Liu^j,
Yalong Zhou^k, Shiqi Tang^l, Kuo Li^m, Ke Yangⁿ, Zheng Yang^o,
Xiaomeng Cheng^p, Ziwan Chen^q, Hua Zhang^r, Chunhu Mo^s

^{a-p}Institute of Geophysical & Geochemical Exploration,
Chinese Academy of Geological Sciences, Langfang, China

^bChina University of Geosciences (Beijing), Beijing, China

^qYunnan Institute of Geological Survey, Kunming, China

^rSichuan Geological Survey, Chengdu, China

^sGuizhou Geological Survey, Guiyang, China

Abstract: The Earth's surface is a complex system involving mutual interactions of its many components, including mountains, rivers, forests, farmlands, lakes and grasses. The interaction and mutual feedback of chemical elements in Earth's surface layer can drive changes in chemical elemental distribution patterns. In this study, we evaluated the mechanisms and interactions driving the distribution patterns of macroelements, probiotics, halogens and heavy metals in soils in Southwest China, based on a systematic geochemical land-quality survey at a scale of 1:250000. The results showed that the parent material determines the natural state of chemical elements in land resources. Epigenetic geochemical dynamics reshapes the distribution patterns of chemical elements in top soil; biogeochemical processes drive the evolutionary trends of land quality; and human activities, such as mining, disrupt the natural evolution of chemical elemental distribution patterns. The establishment of an epigenetic geochemical dynamics theory allows the construction of a framework for understanding the Earth's surface layer and promoting technological innovations for the comprehensive geochemical investigation of land resources.

Keywords: epigenetic geochemical dynamics, driving mechanisms, distribution pattern of elements in soils, Southwest China

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Эпигенетическая геохимическая динамика и движущие механизмы закономерностей распределения химических элементов в почвах Юга-Западного Китая*

© Хансинь Чэн^a, Минь Пэн^b, Чуаньдун Чжао^c, Вэй Хань^d, Хуйянь Ван^e,
Цяолин Ван^f, Фань Ян^g, Фугуй Чжан^h, Чэнвэнь Ванⁱ, Фэй Лю^j,
Ялун Чжоу^k, Шици Тан^l, Ко Ли^m, Кэ Янⁿ, Чжэн Ян^o,
Сяомэн Чэн^p, Цзывань Чэн^q, Хуа Чжан^r, Чуньху Мо^s

^{a-p}Институт геофизических и геохимических исследований

Китайской академии геологических наук, г. Ланфан, Китай

^bКитайский геологический университет (Пекин), г. Пекин, Китай

^cЮньнаньский институт Геологической службы, г. Куньмин, Китай

^dСычуаньская геологическая служба, г. Чэнду, Китай

^eГеологическая служба Гуйчжоу, г. Гуйян, Китай

Резюме: Поверхность Земли представляет собой сложную систему, включающую взаимодействие многих ее компонентов, в том числе гор, рек, лесов, сельскохозяйственных угодий, озер и трав. Взаимодействие и взаимная обратная связь химических элементов в поверхностном слое Земли может привести к изменениям в структуре распределения химических элементов. В этом исследовании авторы оценили механизмы и взаимодействия, определяющие характер распределения макроэлементов, пробитиков, галогенов и тяжелых металлов в почвах Юго-Западного Китая, на основе систематического геохимического исследования качества земли в масштабе 1:250000. Результаты показали, что исходный материал определяет естественное состояние химических элементов земельных ресурсов. Эпигенетическая геохимическая динамика меняет характер распределения химических элементов в верхнем слое почвы, биогеохимические процессы определяют эволюционные тенденции качества земли, а деятельность человека, такая как добыча полезных ископаемых, нарушает естественную эволюционную схему распределения химических элементов. Создание теории эпигенетической геохимической динамики позволяет построить основу для понимания поверхностного слоя Земли и продвигать инновационные технологии для всестороннего геохимического исследования ресурсов земной коры.

Ключевые слова: эпигенетическая геохимическая динамика, движущие механизмы, закономерности распределения элементов в почвах, Юго-Западный Китай

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Background

Epigenetic geochemical dynamics explores the driving mechanisms, dynamic processes, and fractionation mechanisms of the distribution, allocation, migration and evolution of chemical elements and isotopes in surface rocks, water bodies, soils, organisms, atmosphere and other media or layers of the Earth. The Earth's surface is a complex system composed of various interact-

ing components, such as mountains, water bodies, forests, farmlands, lakes and grasslands. This system controls and regulates the natural habitat and maintains the resources required to sustain life on Earth [1]. The pedosphere is the key interface, at which the lithosphere, hydrosphere, biosphere and atmosphere interact. It plays an important role in the redistribution of elements, and is also an essential interface for

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supporting economic and social developments [2]. Presently, the Earth is facing a serious environmental crisis in history. This has caused dysfunction in the Earth's surface layer and led to major changes in the global environment on which human survival depends. Understanding the behavior of chemical elements within the physical, chemical and biological processes of the Earth's surface, along with human-environmental interactions, has inevitably become a hot topic in exploratory geochemistry. Research in this area can reveal the driving mechanisms behind changes in the content and macroscopic distribution of elements in soil, thereby provide scientific solutions for the rational exploitation and utilization of natural resources.

In this study, we analyzed the mountainous regions near several cities including Zhaotong City in Yunnan Province, Bijie City in Guizhou Province, and Yibin and Luzhou Cities in Sichuan Province, and examined the driving mechanisms of chemical elemental distribution in soil.

Geographical and geological settings

The study area is located at the junction of Yunnan, Guizhou and Sichuan Provinces (102–

107° E and 25–30° N). The geographical environment of the area is complex, with different climate types co-existing at different elevations, and with significant regional variations. In the last 45 years, the average annual precipitation of the area was 1,885 mm, while temperature, precipitation and evaporation are increasing [3, 4].

The study area mainly consists of mountains and valleys, showing strong down-cutting and Karst landforms. The terrain slopes northwards, as behind elevation lowers gradually from the southwest to the northeast by a difference of greater than 3,400 m (Fig. 1). Farmlands, forests and grasslands accounted for 48.8 %, 41.4 % and 5.2 % of the total area of the region, respectively, and they represent the major land-use types in the region (Fig. 2).

All strata in the area have different degrees of exposure, where carbonate rocks, such as limestone and dolomite, from various periods, are widely distributed. Basalt, carbonate and sedimentary clastic rocks are the main parent materials of the regional soil (Fig. 3).

Lead (Pb), zinc (Zn) and silver (Ag) constitute the area's dominant mineral resources formed in the Devonian and Permian periods. The ores are

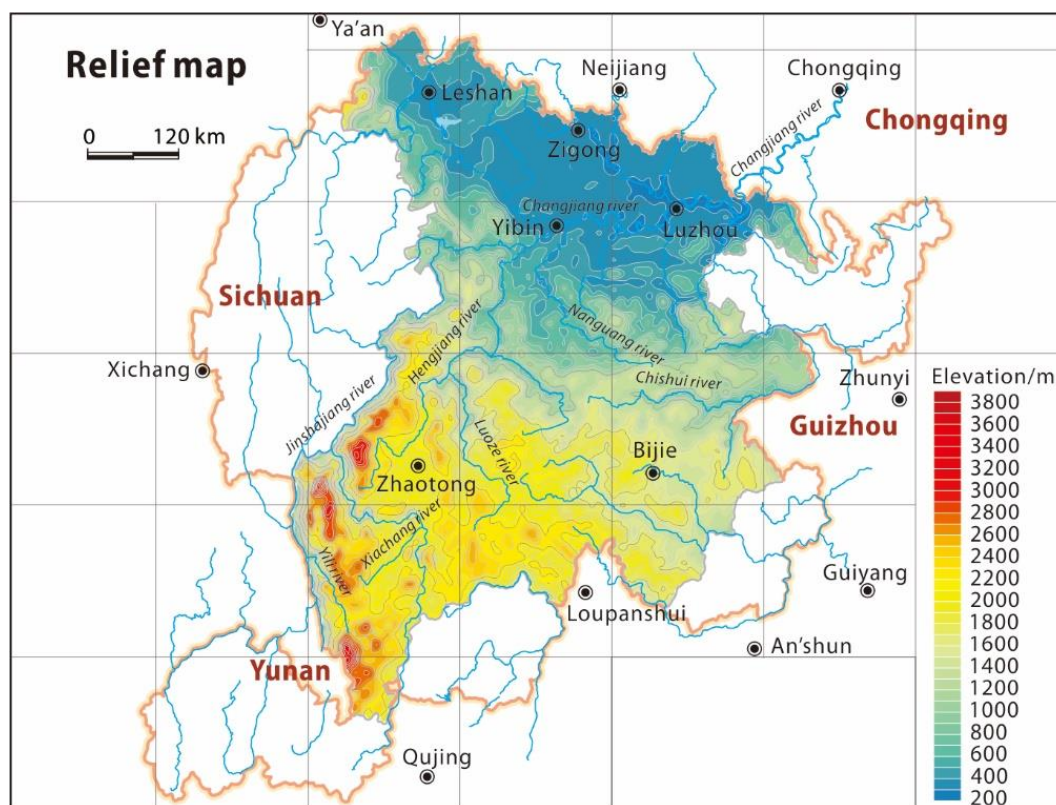


Fig. 1. Terrain distribution in the Zhaotong – Bijie – Yibin – Leshan – Luzhou area

Рис. 1. Распределение высот рельефа в районе Чжаотун – Бицзе – Ибинь – Лэшань – Лучжоу

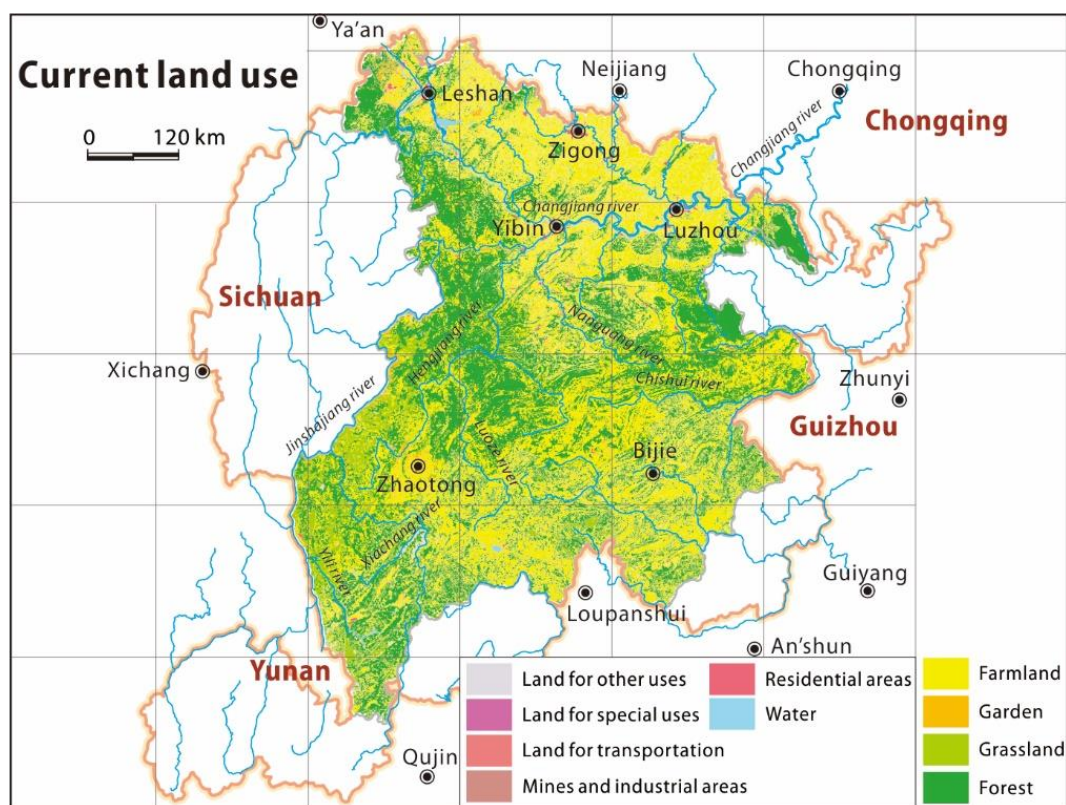


Fig. 2. Land-use types in the Zhaotong – Bijie – Yibin – Leshan – Luzhou area
Рис. 2. Типы землепользования в районе Чжаотун – Бицзе – Ибинь – Лэшань – Лучжоу

usually present in carbonaceous strata, comprising interbedded dolomite, dolomitic limestone, limestone and marl in the area. The area also features paragenetic or associated non-ferrous, rare and precious metal elements such as copper (Cu), iron (Fe), silver, gallium (Ga), cadmium (Cd) and germanium (Ge) [5, 6].

Basalt, carbonate and Pb-Zn-Ag deposits are mainly distributed in the mid- and high-elevation mountainous areas above 1,000 m (Fig. 3). The Emeishan large igneous province (LIP) resulted from a mantle-plume melting event spanning less than 1 Ma at approximately 259 Ma [7]. Spatially, Emeishan basalt is mainly distributed in Yunnan, Sichuan and Guizhou Provinces along the western margin of the Yangtze Craton. This region extends to Guangxi and Chongqing in the east and is bordered by the Ailaoshan – Red River fault to the west and the Longmenshan – Xiaojiang River fault to the northwest. As it is near the Sanjiang tectonic belt, the Emeishan LIP's complex

geological history includes strong deformation and destructive events that resulted in approximately $0.25 \cdot 10^6$ km² exposed area. The basalt in the study area is located within the continental rift system in the Emeishan basalt zone [8], where the exposed area covered approximately 8,867 km² (Fig. 3) or approximately 10.46 % of the total study region; while the exposed carbonate rocks covered 20,934 km² or approximately 24.70 % of the study area.

The Geochemical Atlas of China^{1,2} and the Geochemical Survey Report of Chinese Farmland³ clearly show abnormal enrichment of metal elements in the soil and stream sediments in the Emeishan basalt and the southwestern carbonate areas. These areas have high metallic elemental contents relative to elsewhere in China.

Therefore, it is essential to study the epigenetic geochemical processes in these areas and the impact of Pb and Zn mining activities on the regional elemental distribution. The findings

¹ Xie X., Ren T., Sun H. Geochemical atlas of China. Beijing: Geological Publishing House, 2012. 120 p.

² Xie X., Cheng Z., Zhang L., et al. Geochemical atlas of 76 elements in Southwest China. Beijing: Geological Publishing House, 2008. 219 p.

³ China Geological Survey. Geochemical survey report on China's cultivated land (2015). Available from: <http://www.cgs.gov.cn/upload/201506/20150626/gdbg.pdf> [Accessed 25 June 2015].

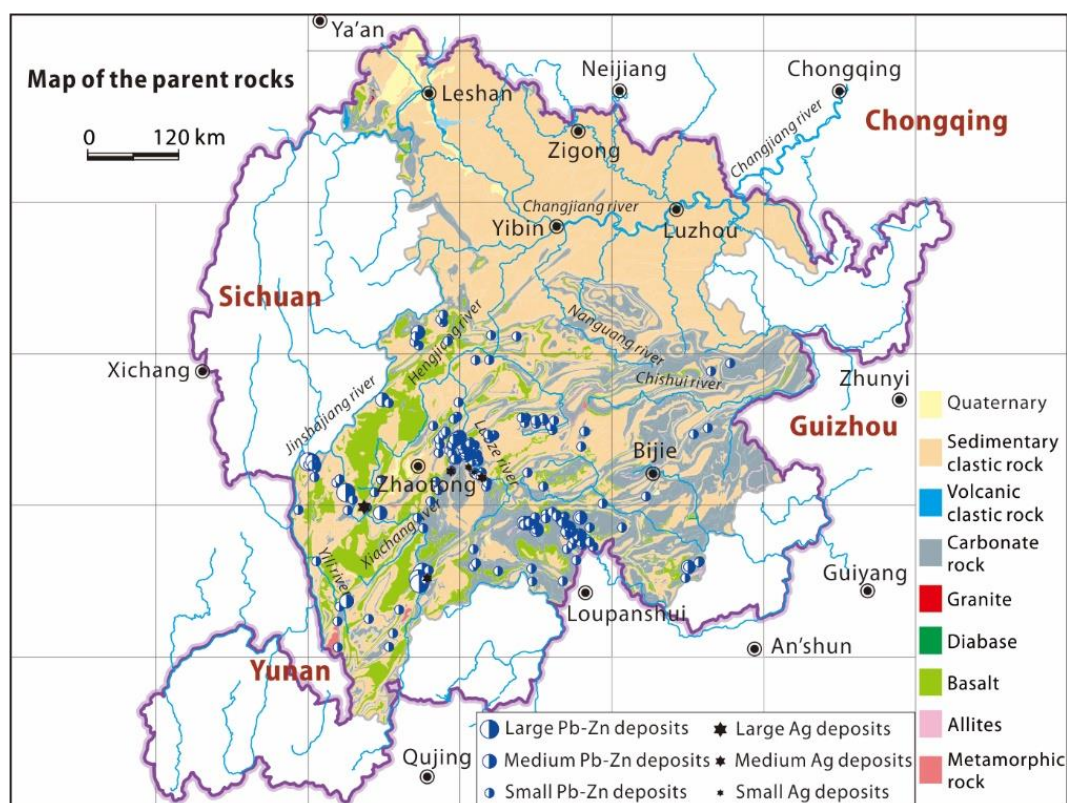


Fig. 3. Distribution map of the parent materials and Pb-Zn-Ag deposits in the Zhaotong – Bijie – Yibin – Leshan – Luzhou area

Рис. 3. Карта распределения материнских горных пород и месторождений Pb-Zn-Ag в районе Чжаотун – Бицзе – Ибинь – Лэшань – Лучжоу

can facilitate the advancements of the theories of epigenetic geochemical dynamics, as well as development of geochemical surveys and monitoring systems for land resources, and applications of exploratory geochemistry in natural resource management.

Data sources

The data used in this study were obtained through tests in accordance with the “Multi-Purpose Regional Geochemical Survey” specifications (1:250,000) (DZ/T 0258-2014)⁴. The sampling depths of the top and deep soil samples were 0–20 cm and 150–180 cm, respectively. The samples were analyzed in the Central Laboratory of the Institute of Geophysical and Geochemical Exploration, Chinese Academy of Geological Sciences. The tested elements and indicators included Ag, As, Au, B, Ba, Be, Bi, Br, Cd, Ce, Cl, Co, Cr, Cu, F, Ga, Ge, Hg, I, La, Li, Mn, Mo, N, Nb, Ni, P, Pb, Rb, S, Sb, Sc, Se, Sn, Sr,

Th, Ti, Tl, U, V, W, Y, Zn, Zr, SiO₂, Al₂O₃, TFe₂O₃, MgO, CaO, Na₂O, K₂O, TC, organic carbon (C_{org}) and pH. The schemes for analysis and quality control used in this study are as described in the references [9]. A quality assessment of the data, analyzed according to the prescribed procedures, showed that the data are reliable and passed the acceptance and database audits.

Mechanisms driving the macroscopic distribution of major elements

During complex physical and chemical weathering processes, soil parent materials can form in-situ residual soil. Alluvial and proluvial soils may also appear in downstream areas due to transportation and sedimentation processes when slopes and rivers are present. Deep soil is less affected by industrial and agricultural activities, in which elemental distribution is mainly controlled geologically. Typically, when there is no significant impact from human activities, the

⁴ Ministry of Land and Resources of the People's Republic of China. Code for multi-target regional geochemical surveys (1:250000) (DZ/T 0258-2014). Beijing: China Standard Press, 2015. 43 p.



elemental distribution patterns of the top soil reflects that of the deep soil. However, owing to the extensive and frequent contacts between the top soil and atmosphere, water bodies, organisms or human beings, the elemental distribution patterns in soil can be considerably modified. Emeishan basalt and carbonate rocks are the two most common soil parent materials in the study area. The dynamic processes of surface erosion, transportation and sedimentation, caused by marked elevation changes and biogeochemical effects of extensive forests, are a starting point for characterizing the mechanisms driving the macroscopic distribution of elements in this region.

Main weathering and soil formation processes of basalt and carbonate rocks. Emeishan basalt is mainly composed of basic plagioclase and pyroxene minerals with some olivine, amphibole and biotite. In an epigenetic environment, the physical weathering processes drive basic feldspar ($\text{Na}[\text{AlSi}_3\text{O}_8] - \text{Ca}[\text{Al}_2\text{Si}_2\text{O}_8]$) to fracture along the cleavage planes. Further, the chemical weathering processes cause basic feldspar decomposition, i.e., basic feldspar \rightarrow hydromica + K^+ , Na^+ , $\text{Ca}^{2+} \rightarrow$ kaolinite (montmorillonite) + $\text{SiO}_2 \rightarrow$ opal + gibbsite, leading to loss of K^+ , Na^+ , Ca^{2+} and H_2SiO_4 . Olivine ($(\text{Mg}, \text{Fe})_2\text{SiO}_4$) and pyroxene ($\text{Ca}(\text{Mg}, \text{Fe}, \text{Al})[\text{Si}, \text{Al}]_2\text{O}_6$) are unstable Fe and Mg minerals that are highly susceptible to weathering. During weathering and soil formation, especially under the action of carbonic acid, Ca^{2+} , Mg^{2+} and Fe^{2+} ions are first separated to form bicarbonates during decomposition, then dissolved in water and carried away. Under oxidation, the low-valent iron

in these minerals is oxidized to high-valent iron, forming hydrous iron oxides. These minerals remain in-situ to form red-, brown-, and hazel-colored soils.

The main mineral component of carbonate rocks is calcium carbonate. Under the long-term action of CO_2 -rich rainwater, calcium carbonate undergoes chemical dissolution ($\text{CaCO}_3 + \text{H}_2\text{O} + \text{CO}_2 \rightarrow \text{Ca}^{2+} + 2\text{HCO}_3^-$). Overall, 90 % of Ca^{2+} , Mg^{2+} and other dissolved substances are lost due to water transportation. The remaining materials that are insoluble in acids, such as quartz, feldspar, clay and Fe and Al oxides, remain on the surface to form soil. Therefore, the time used by the carbonate rock parent materials to form soil is up to 10 times that used by other parent materials [10, 11].

Basalt is located in the continental rift system in the Emeishan basalt area. Compared to its parent rock, the soil that basalt forms was rich in Al and Fe but depleted in Ca, Mg, Na, K and Si (Table 1). During soil formation from carbonate parent materials, approximately 96 % of Ca and 52 % of Mg were lost, whereas Fe, Al, Si, K and Na were enriched by 164, 5.5, 4, 16 and 2.9 times, respectively (Table 2).

Mechanisms driving the macroscopic distribution of Fe and Al. The above-mentioned enrichment and depletion characteristics of major elements during soil formation have been clearly verified through their spatial distributions. The deep and top soils above the basalt and limestone parent materials were considerably enriched in Fe and Al, indicating significant control by soil parent materials over the distributions

Table 1. Chemical compositions of major elements in Emeishan basalt and soil

Таблица 1. Химический состав основных элементов в Эмейшанских базальтах и почвах

Statistics	$w_B / \%$						
	SiO_2	Al_2O_3	TFe_2O_3	MgO	CaO	Na_2O	K_2O
Number of samples	859	859	859	859	859	859	859
Minimum	27.83	9.4	6.48	0.32	0.09	0.06	0.09
Maximum	67.47	25.39	31.41	8.73	14.04	3.2	3.5
Mean	42.17	17.16	17.4	1.14	0.73	0.39	1.08
Median	41.6	17.56	17.97	1.01	0.52	0.29	1.04
Standard deviation	6.43	2.28	3.26	0.6	0.94	0.35	0.36
Average chemical composition of the Emeishan basalt ($n = 416$)*	50.03	13.89	14.06	5.17	8.45	2.84	1.33
Soil / Rock	0.84	1.24	1.24	0.22	0.09	0.14	0.81

Note. * – see reference [8].

Примечание. * – см. источник [8].



Table 2. Chemical compositions of major elements in carbonate rocks and soil
Таблица 2. Химический состав основных элементов в карбонатных породах и почвах

Sampling medium	Statistics	w _B / %						
		SiO ₂	Al ₂ O ₃	TFe ₂ O ₃	MgO	CaO	Na ₂ O	K ₂ O
Carbonate	Number of samples	34	34	34	34	34	34	34
	Minimum	9.75	1.9	0.01	0.03	0.1	0.02	0.01
	Maximum	96.21	4.66	0.43	7.23	55.39	0.06	1.31
	Mean	16.13	2.62	0.05	0.93	48.33	0.04	0.06
	Median	10.03	2.49	0.01	0.37	52.19	0.04	0.01
	Standard deviation	20.34	0.48	0.1	1.43	12.89	0.01	0.23
Soil	Number of samples	495	495	495	495	495	495	495
	Minimum	30.72	5.88	1.79	0.13	0.12	0.05	0.17
	Maximum	87.18	26.76	24.48	1.33	22.13	0.73	3.99
	Mean	64.34	14.36	7.54	0.45	1.99	0.12	1.01
	Median	63.99	14.24	6.81	0.43	0.37	0.09	0.78
	Standard deviation	12.68	4.3	3.82	0.2	3.9	0.09	0.7
Soil / Rock		3.99	5.47	164.3	0.48	0.04	2.87	16.01

of Fe and Al (Figs. 4 and 5). Furthermore, Fe and Al contents in soil increased with elevation (Fig. 6), suggesting that during soil formation, Fe and Al did not substantially migrate from high elevations to the base of mountains. Therefore, the high Fe and Al contents in soil were the result of secondary enrichment due to parent material weathering.

The top soil is in direct contact with the atmosphere and biosphere and affected by industrial and agricultural activities. During soil formation, the soluble base cations in the top soil are more likely to be washed away, resulting in lower pH (5.82) than the deep soil (6.04). Additionally, irrespective of ground cover types (farmlands, forests, grasslands, etc.), the root system of vegetation is mainly concentrated at a shallow depth of tens of centimeters. Moreover, the root and microbial growth activities of crops can produce large quantities of organic acids, adding to the already highest levels of acid rain in China the study area receives. In acidic environments, Fe²⁺ and Fe³⁺ are formed during the weathering of Fe rich ferromagnesian minerals. The migration capacity of Fe²⁺ is much greater than that of Fe³⁺, and exchangeable Al is more likely to enter the soil solution and migrates with water. Together, these factors were suggested as the main mechanism driving the lowering of TFe₂O₃ and Al₂O₃ concentrations in the top soil, in this case, by 4.6 % and 4.9 %, respectively (Fig. 6), compared to the top soil.

Mechanisms driving the macroscopic distributions of Na, K, Ca, Mg and Si, and pH. Because Na, K, Ca and Mg are soluble in water, these elements are prone to being washed away and can easily enter the epigenetic environment during weathering and soil formation. The levels of some elements such as Ca, Na (Fig. 7), K and Mg (not shown) in the top and deep soils increased rapidly with decreasing elevations (Fig. 7), suggesting that these elements mainly entered the surface water system and migrated from high- to low-elevation areas under the action of hydrodynamic forces.

Owing to their different geochemical properties, the above-mentioned elements have significantly different migration and transportation pathways after entering the epigenetic environment. Although Na and K have similar geochemical properties, the hydration energy of Na (indicating how closely the ions are surrounded by water molecules) is greater than that of K. A larger hydration energy means the ion is more difficult to be adsorbed. Therefore, in an epigenetic environment, Na is more likely to migrate with water, whereas K is more easily adsorbed by clay minerals or colloids. This is also the main reason Na was concentrated near the lowest terrain along the Yangtze River (Fig. 8, a), whereas K only occurred in low concentrations in the same area (Fig. 8, b).

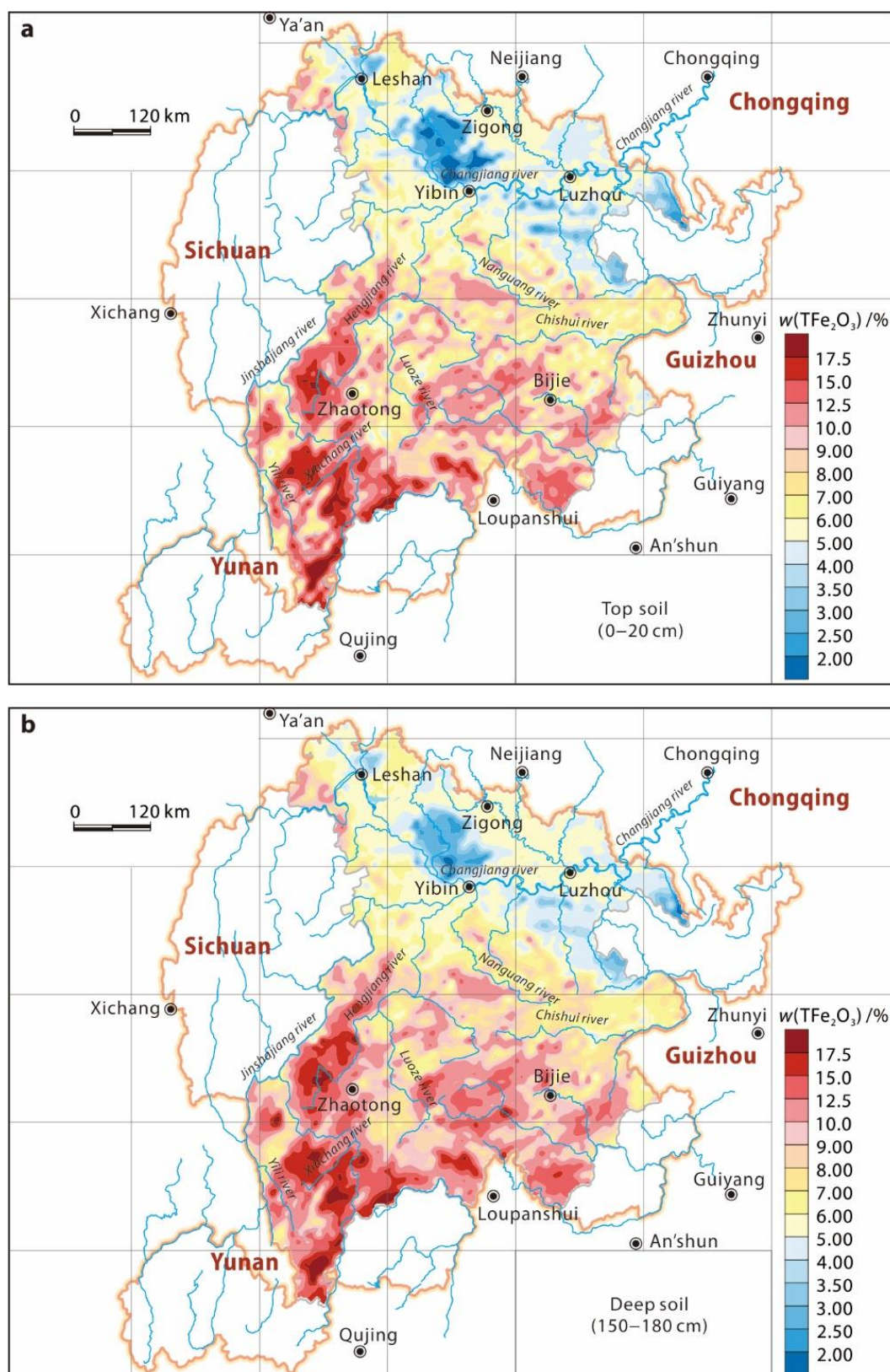


Fig. 4. The TFe_2O_3 distribution pattern in top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area

Рис. 4. Схема распределения TFe_2O_3 в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах района Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

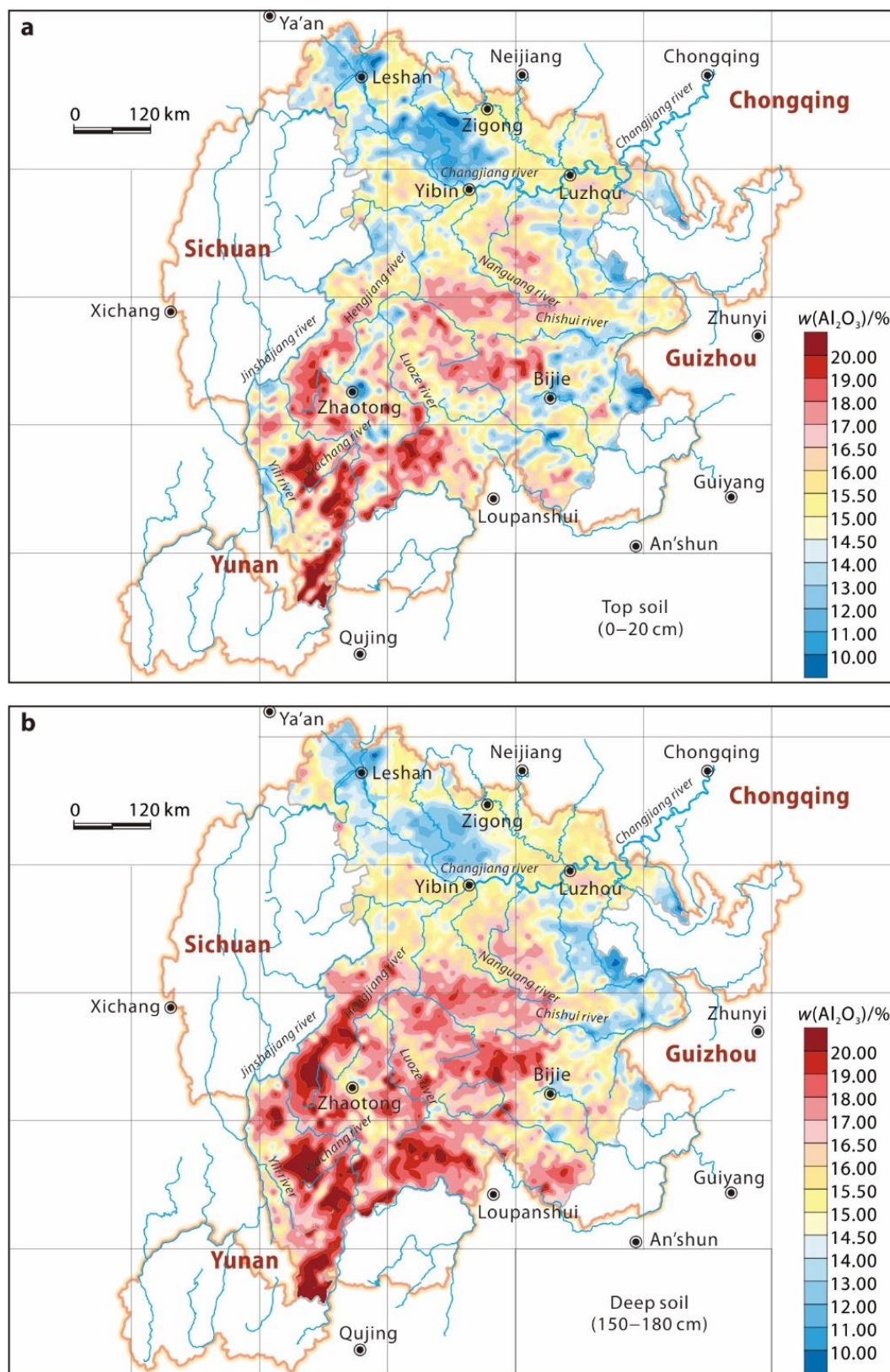


Fig. 5. The Al_2O_3 distribution pattern in top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 5. Схема распределения Al_2O_3 в верхних (0–20 см) (а) и глубоких (150–180 см) (б) почвах района Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

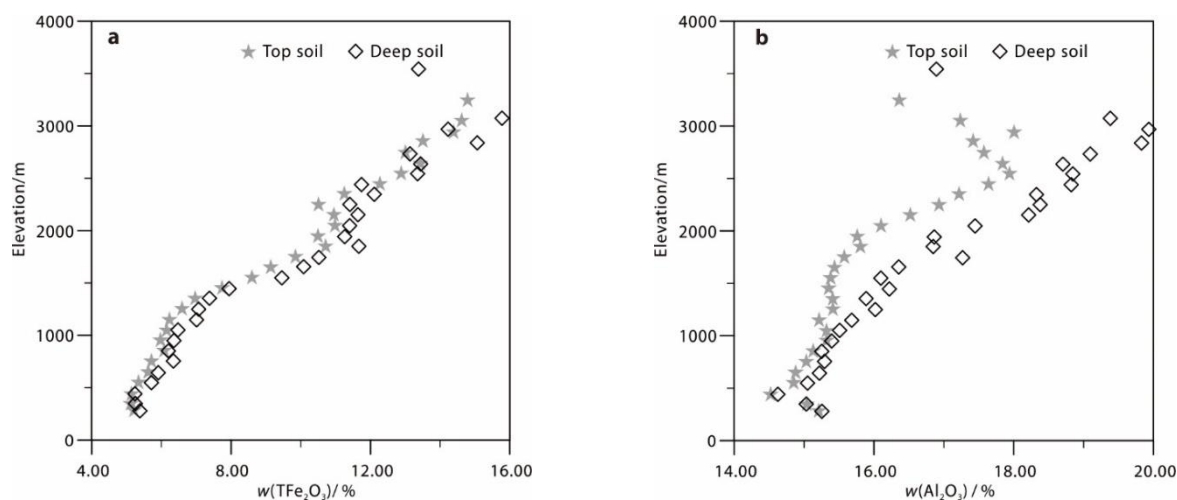


Fig. 6. Plot of TFe_2O_3 (a) and Al_2O_3 (b) contents in top and deep soils vs. elevation in the Zhaotong – Bijie – Yibin – Luzhou – Leshan area

Рис. 6. График содержания TFe_2O_3 (a) и Al_2O_3 (b) в верхних и глубоких почвах в зависимости от высоты в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

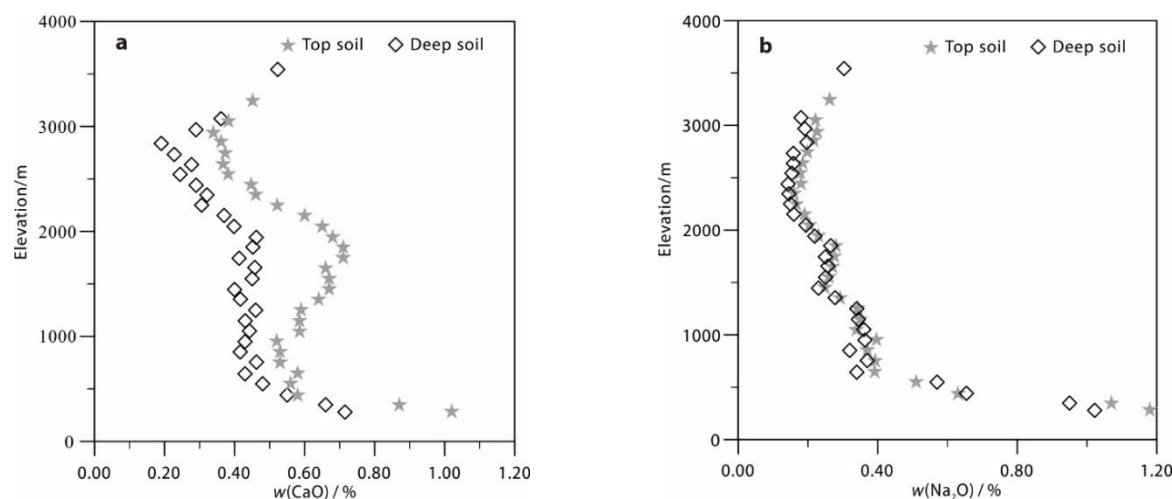


Fig. 7. Plot of CaO (a) and Na_2O (b) contents in top and deep soils vs. elevation in the Zhaotong – Bijie – Yibin – Luzhou – Leshan area

Рис. 7. График содержания CaO (a) и Na_2O (b) в верхних и глубоких почвах в зависимости от высоты в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

The solubilities of sodium chloride and sodium hydroxide at room temperature were 36 g/NaCl and 109 g/NaOH in 100 g water, respectively, while that of calcium bicarbonate was 16.6 g/ CaHCO_3 in 100 g water. These compounds appeared mainly in the forms of Ca^{2+} , Cl^- , Na^+ and OH^- in natural water. As temperature rises, Ca^{2+} precipitates before Na^{2+} . All Na, K, Ca and Mg were lost and enter into the water system during weathering and soil formation. However, due to solubility differences, Ca^{2+} and Na^+ , Ca and Mg (not shown) were mainly concentrated in the soils along both sides of the tributary systems, e.g., the Jinsha and Chishui Rivers, by the action of hydrodynamics (Fig. 9, a). Contrarily, Na migrated over a long distance and concentrated within the

lowest-lying areas near the Yangtze River (Fig. 8, a).

Potassium (K^+), Na^+ , Ca^{2+} , Mg^{2+} and other basic ions can all neutralize H^+ and prevent and regulate the acidification of water bodies and soil. Weathering and soil formation of the widely distributed basalt and carbonate rocks provide sufficient basic ions, such as K^+ , Na^+ , Ca^{2+} and Mg^{2+} , to the surface water for buffering acidity and maintaining moderately alkaline conditions in the soils along both sides of the water systems. Figure 9, b shows that soils on both sides of the Yangtze River tributaries, including the Jinsha, Hengjiang, Nanguang and Chishui Rivers, were moderately alkaline. These regions overlap precisely with the Ca rich areas, a clear indication of H^+ neutralization by Ca^{2+} and other basic ions.

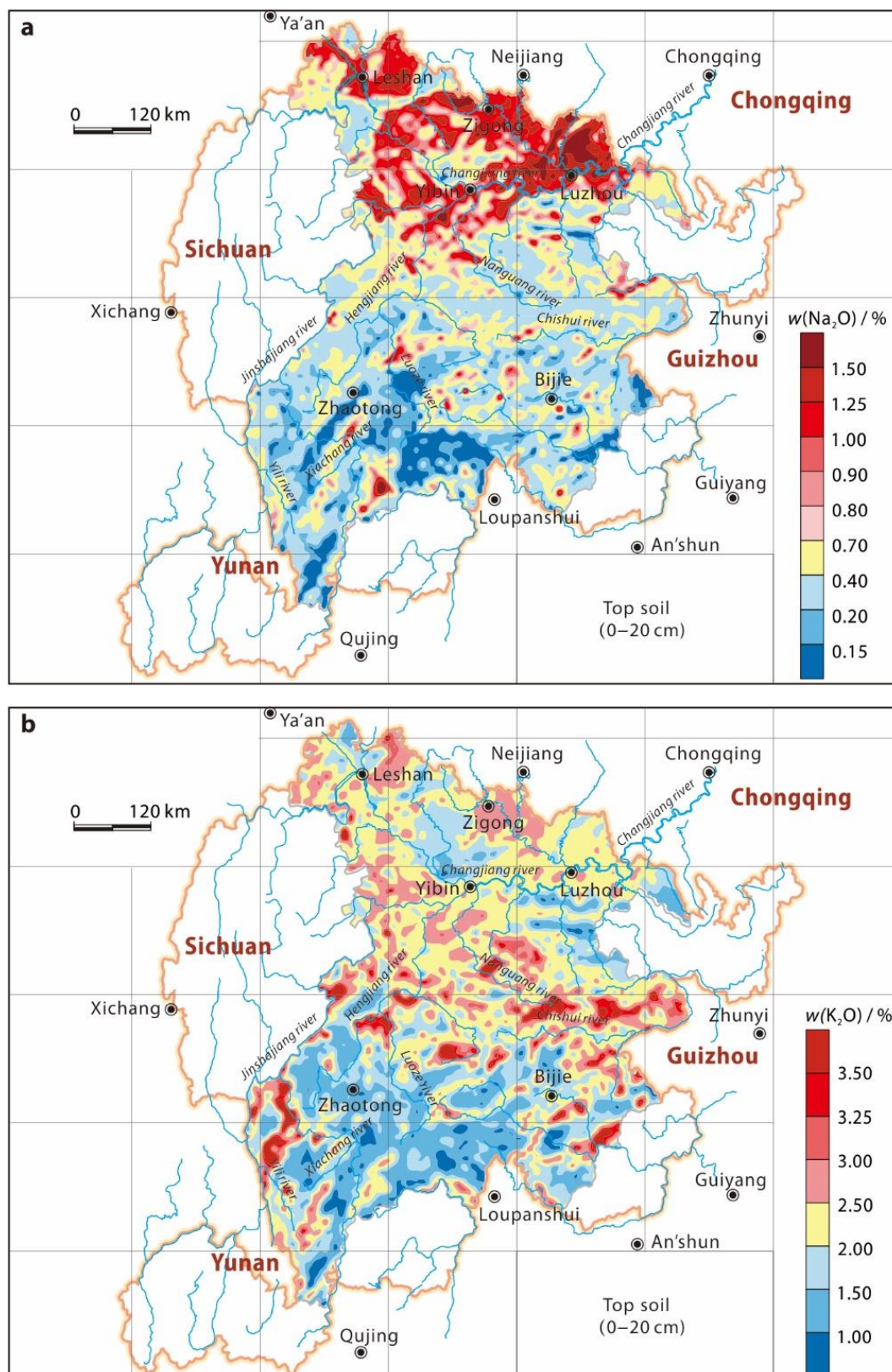


Fig. 8. Distribution pattern of Na_2O (a) and K_2O (b) in the top (0–20 cm) soil of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 8. Схема распределения Na_2O (a) и K_2O (b) в верхних (0–20 см) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

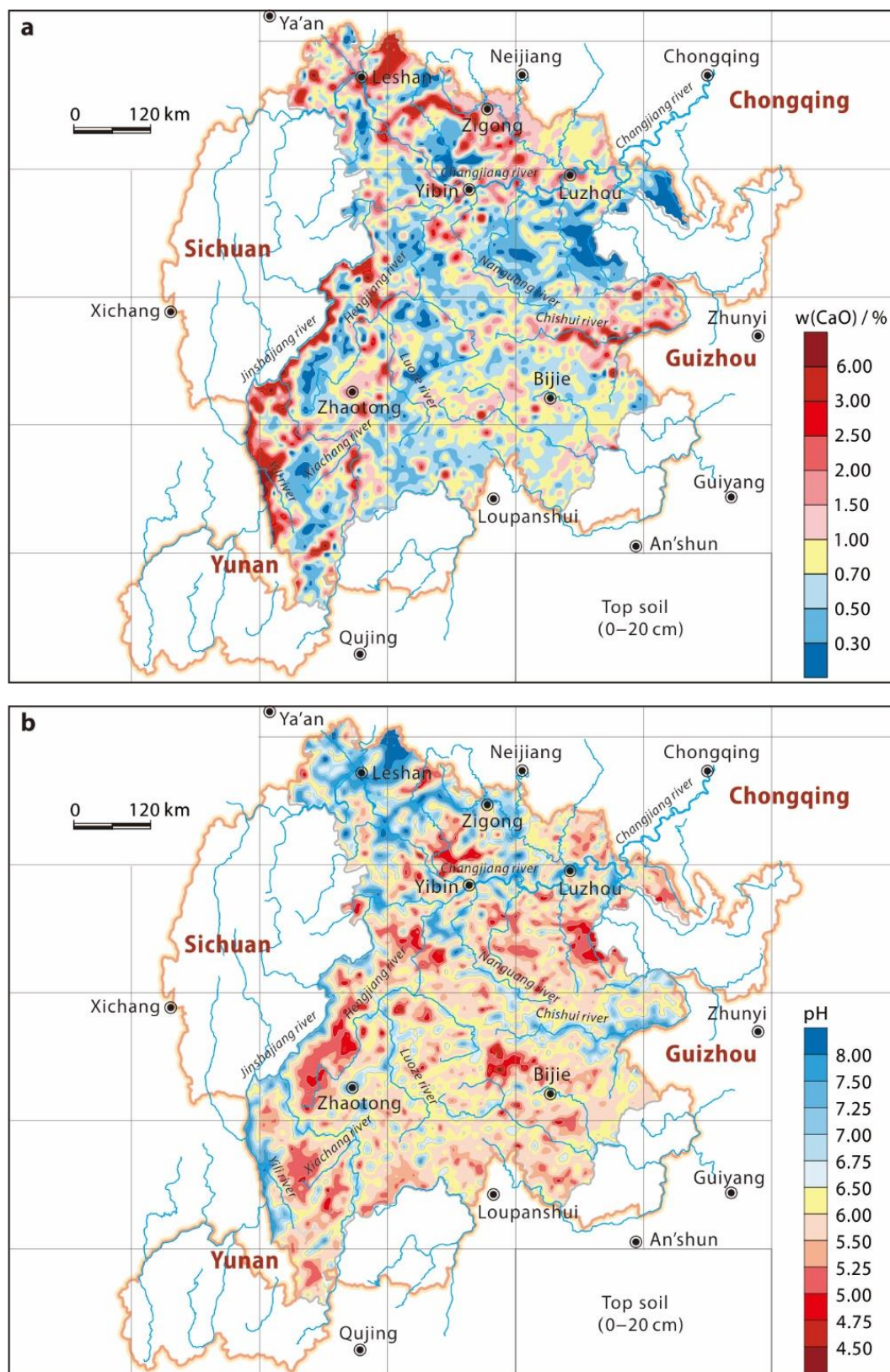


Fig. 9. Distribution pattern of CaO (a) and pH levels (b) in the top (0–20 cm) soil of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 9. Схема распределения CaO (a) и уровней pH (b) в верхних (0–20 см) почвах района Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань



The insoluble materials formed via weathering and soil formation from carbonate rocks mainly include extremely fine-grained quartz, feldspar and clay [10, 11]. The study area has abundant annual rainfall, and the terrain exhibits strong undercutting. Silicone content increased

rapidly as elevation decreases (Fig. 10), indicating that, under the action of epigenetic hydrodynamics, insoluble minerals, such as quartz and feldspar, migrated along the water system and deposited in the lower-lying terrain in the Leshan – Yibin – Luzhou area (Fig. 11).

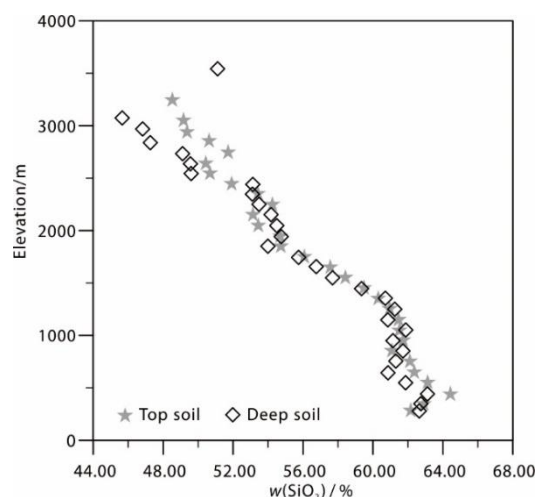


Fig. 10. Plot of SiO_2 content in top and deep soils vs. elevation in the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 10. График содержания SiO_2 в верхних и глубоких почвах в зависимости от глубины в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

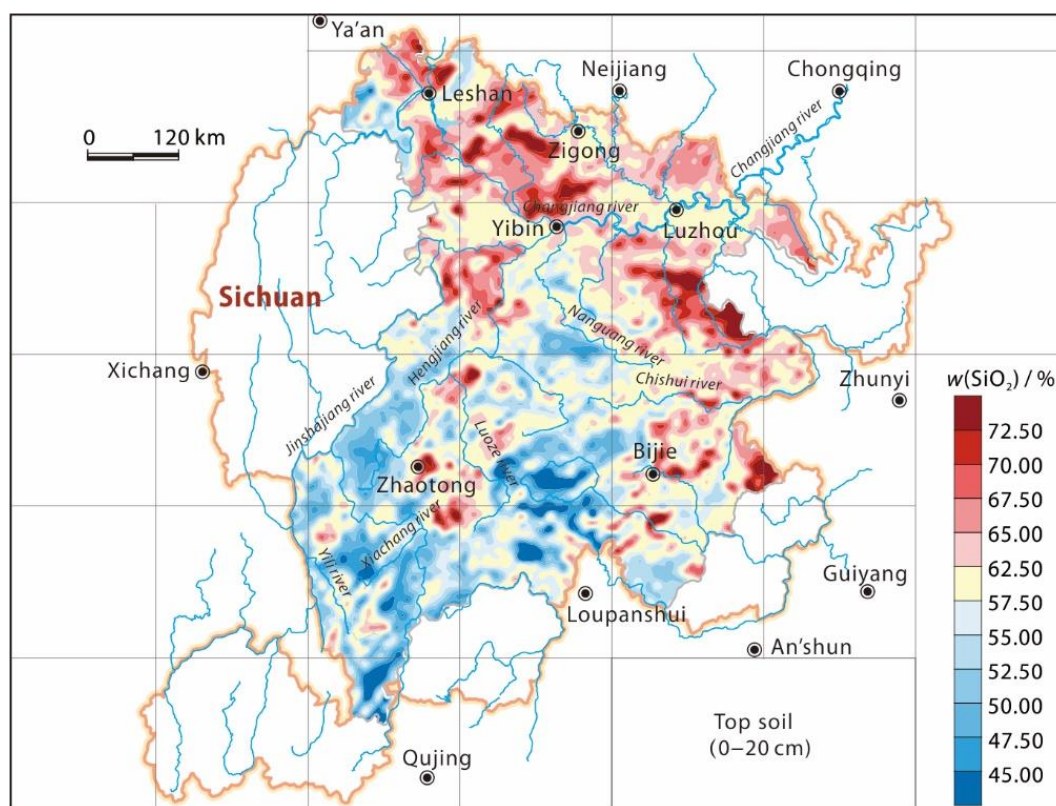


Fig. 11. Distribution pattern of SiO_2 in the top (0–20 cm) soil in the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 11. Схема распределения SiO_2 в верхних (0–20 см) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань



Mechanism driving the macroscopic distribution of C_{org}, N, S and P. According to the procedures of the National Land Quality Geochemical Survey, we analyzed the levels of 52 elements, pH and C_{org}⁵. Thirty-five of the elements participate in biogeochemical processes and have clear biological effects. They included Na, K, Be, Mg, Ca, V, Cr, Mo, W, U, Mn, Fe, Co, Ni, Cu, Ag, Zn, Cd, Hg, B, Al, Ti, C, Si, Sn, Pb, N, P, As, Sb, S, Se, F, Cl and I [12]. In particular, C, N, P and S were the main elements involved in the biogeochemical cycle.

In the study area, the C_{org}, N, S and P contents in the top soil were significantly greater than in the deep soil. As elevation increases, the elemental contents in both soils gradually in-

creased. The absolute increase of the four elements in the top soils of the mid- and high-elevation mountainous areas above 2,000 m were significantly greater than that of the low-elevation areas (Fig. 12). In the major agricultural production region, the top soil was only slightly enriched in all four elements. Organic carbon (Fig. 13), N (Fig. 14), S and P (not shown) were significantly enriched in the top soils of the Zhaotong – Bijie forest and grassland areas above 2,000 m.

Carbohydrates, lipids, proteins and nucleic acids are the four basic substances essential for life; and C, O, H, N, P and S are the major elements required for the synthesis of these basic substances. In the biogeochemical process of plant growth, C and N are mainly involved in the

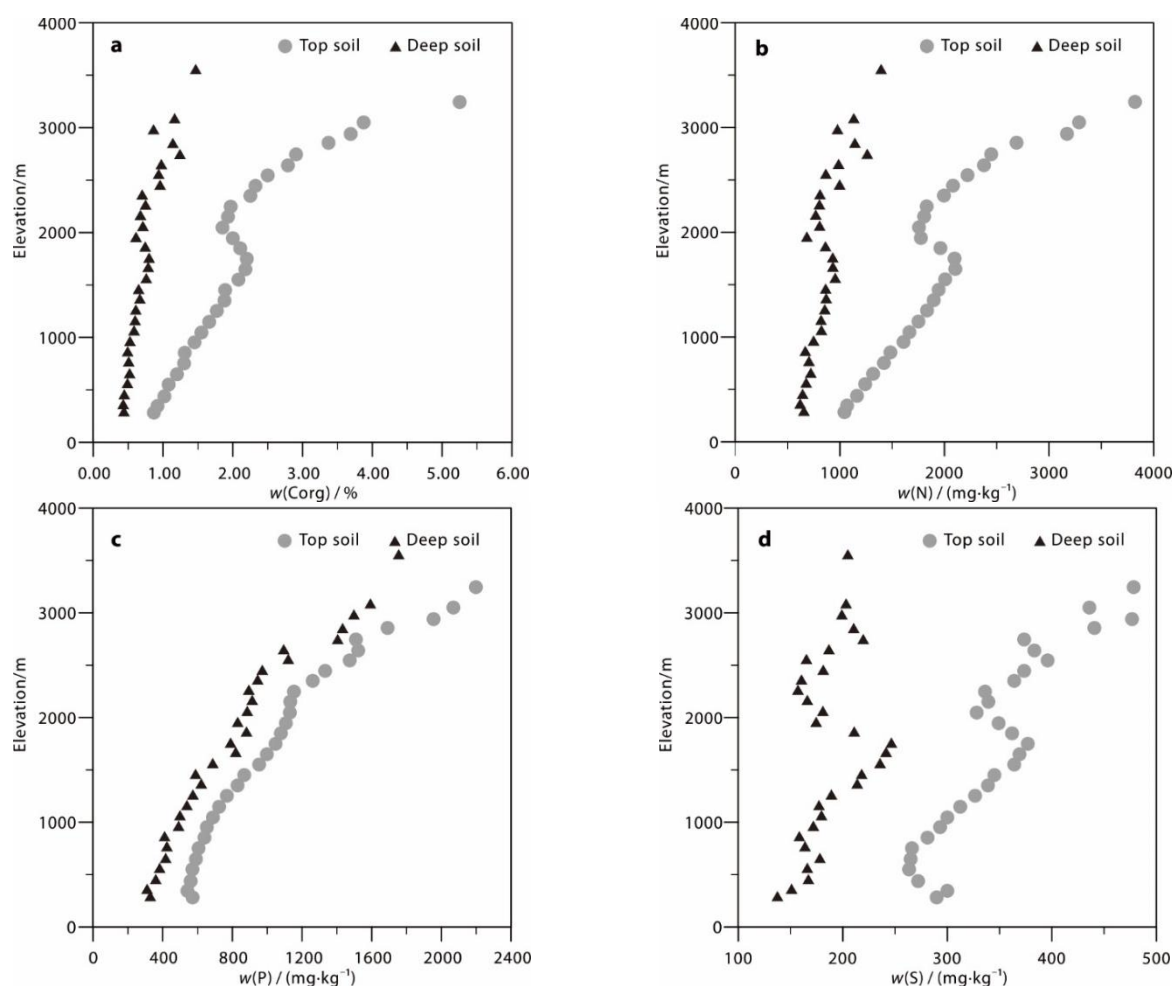


Fig. 12. Plot of C_{org} (a), N (b), P (c) and S (d) contents in the top and deep soils vs. elevation in the Zhaotong – Bijie – Yibin – Luzhou – Leshan area

Рис. 12. График содержания C_{org} (a), N (b), P (c) и S (d) в верхних и глубоких почвах в зависимости от глубины в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

⁵ Ministry of Land and Resources of the People's Republic of China. Code for multi-target regional geochemical surveys (1:250000) (DZ/T 0258-2014). Beijing: China Standard Press, 2015. 43 p.

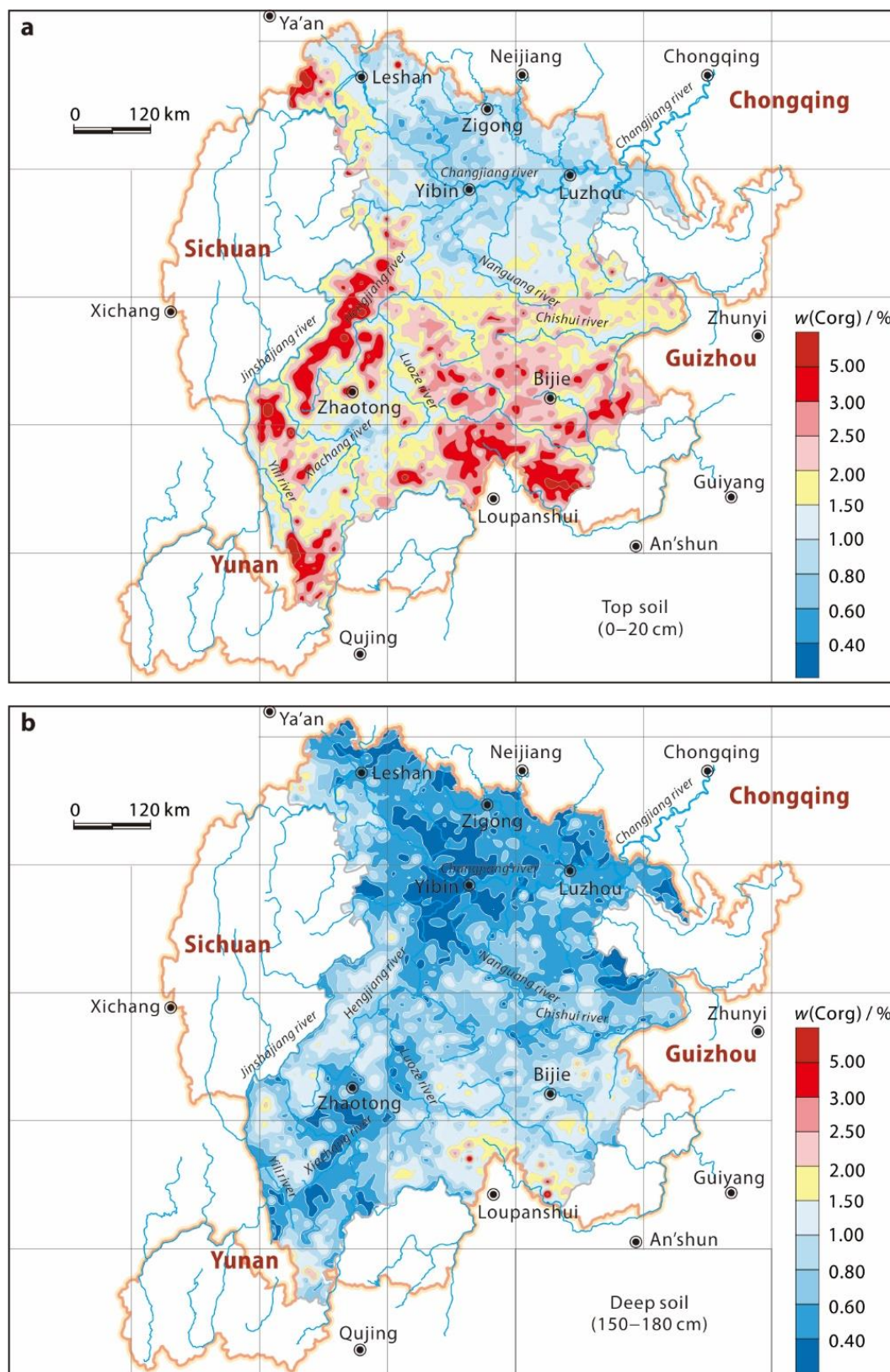


Fig. 13. Distribution pattern of C_{org} in the top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 13. Схема распределения C_{org} в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

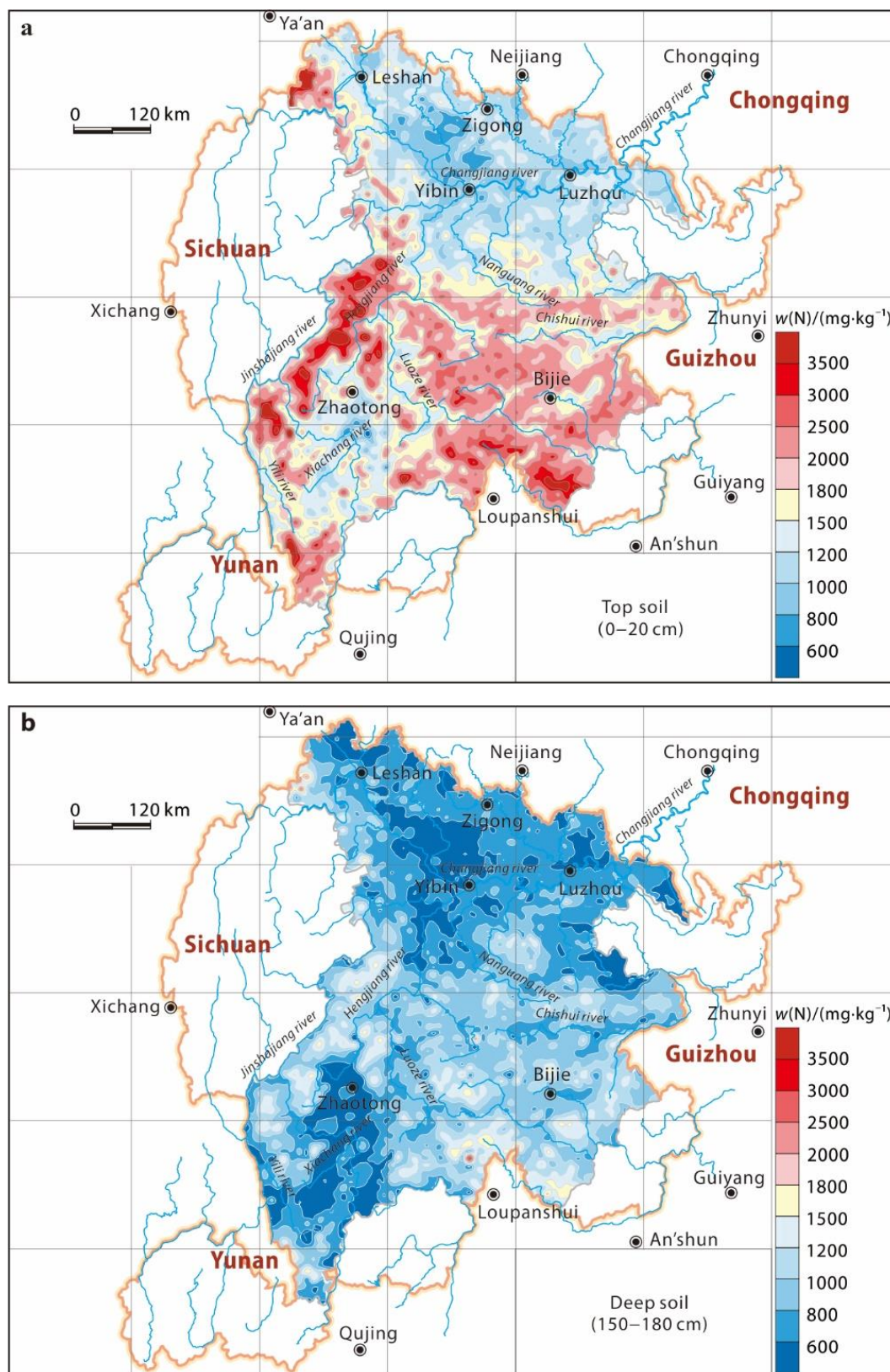


Fig. 14. Distribution pattern of N in the top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 14. Сема распределения N в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань



synthesis of carbohydrates and lipids, while S and P are needed to synthesize proteins and nucleic acids. Thus, the C/N ratio remains fixed in the plant body (C:N = 15). The functions of C, N, S and P render them to follow nearly identical evolutionary patterns in plant production. Although a large amount of chemical fertilizers (mainly nitrogen fertilizers) have been used in agricultural production in China in the last 30 years [13], analyses clearly demonstrate that C_{org}, N, S and P enrichment in the top soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area is driven by the biogeochemical processes of forest and grassland ecosystems. Despite minor cumulation of C_{org}, N, S and P in the top soils of the Yibin – Luzhou – Leshan agricultural area, the accumulation effects attributed to intensive agricultural activities are significantly smaller than those naturally occurring in forest and grassland ecosystems.

Mechanism driving the macroscopic distribution of Cl, Br and I

The Cl, Br and I concentrations in the top soils of the study area were 23.3 %, 51.1 % and 23.5 %, respectively, higher than that in the deep soil; however, the F content was slightly lower in the top soil (-2.8 %).

The combined analyses of elevation variations and the F, Cl, Br and I contents in the top and deep soils showed that the F content was basically the same at different elevations, but highest at around 1,500 m above sea level, indicating that F does not change significantly with elevation in an epigenetic environment (Fig. 15, a). Hence, F concentrations showed consistent spatial distribution patterns in both soils (not shown). Moreover, F rich areas coincided with the carbonate rich areas. Fluorine distribution is mainly controlled by the carbonate parent materials.

Fluorine, Cl, Br and I are halogen elements known by their high chemical reactivity. Chlorine often exists as a monovalent anion (Cl⁻) as well as Cl(V) and Cl(VII) species in the natural environment. Chlorine released during soil formation is mostly in water-soluble form and eventually migrates into the ocean along with water. Therefore, the Cl contents in marine sediments or sedimentary rocks are relatively high. We found that Cl, Br and I levels in the top soil were all significantly

higher than in the deep soil. However, chlorine content increased with decreasing elevation (Fig. 15, b), indicating Cl migration from medium and high elevations to the low hilly areas, whereas Br and I contents increased with increasing elevation (Fig. 15, c,d).

The Cl distribution pattern showed that it had significantly enriched the top soils on both sides of the Yangtze River and in the Pd-Zn-Ag deposit in the Leshan – Yibin – Luzhou area (Fig. 16), consistent with extensive distribution of Cl rich carbonate rocks in the study area. Chlorine released during weathering and soil formation is concentrated along both sides of the Yangtze River, due to the action of hydrodynamic forces, and forms an ionic compound with Na. This is the primary reason we detected abnormally high levels of Cl and Na (Fig. 8, a) on both sides of the Yangtze River.

The Cl anomaly region between the Luoze and Xiaochang Rivers is near a Pd-Zn-Ag mine cluster (Fig. 3). A study on the mineral composition of the deposits found that chlorargyrite exists in the oxidation zone of the large Pb-Zn-Ag deposits in Yinchangpo, Guizhou Province [14]. The high Cl content in chlorargyrite (24.7 %) suggested that natural weathering and mining activities might have driven the high degree of Cl enrichment in the deposits.

Due to the impact of the parent materials, the deep soil above the basalt and carbonate source rocks were generally enriched in Br and I (Figs. 17, b and 18, b). In the top soil, especially in mid-alpine forests with basalt parent materials, Br and I exhibited distinct cumulative distribution patterns (Figs. 17, a and 18, a).

In the natural environment, Br and I contents in the soil are usually much higher than in the parent rocks. According to classical geochemical theories, soil Br and I are believed to mainly come from the atmosphere [15], while atmospheric Br and I originate from ocean evaporation [16]. We found the soil contents of both elements gradually increased with increasing elevation, especially above 2,000 m, indicating high mountain blockage and abundant rainfall are the main reasons for the high Br and I soil contents. Bromine and I in the natural environment mainly exist as compounds. Alpine terrain and precipitation scouring can enable iodine compounds to enter water

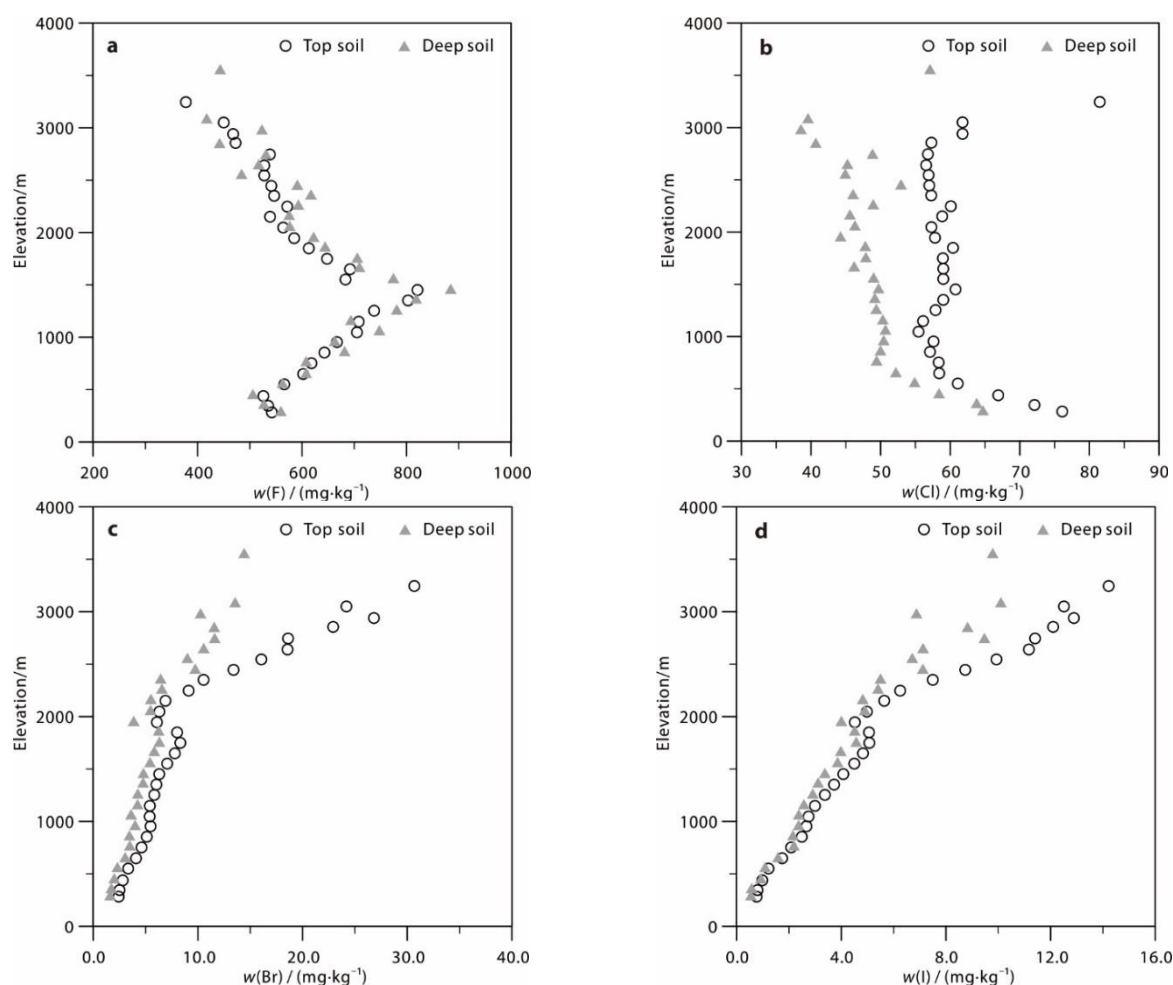


Fig. 15. Plots of F (a), Cl (b), Br (c) and I (d) contents in the top and deep soils vs. elevation in the Zhaotong – Bijie – Yibin – Luzhou – Leshan area

Рис. 15. График содержания F (a), Cl (b), Br (c) и I (d) в верхних и нижних почвах в зависимости от глубины в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

systems and flow into the ocean. However, the increasing Br and I contents in the top soil of the Zhaotong – Bijie alpine valley and low Br and I contents in the soils along the Yangtze River (Fig. 15, c,d), clearly indicated that Br and I do not exist as water-soluble compounds in soil.

A literature review suggests that sediments rich in organic matter are strongly enriched with iodine; such relationship can be used to determine iodine-holding ability of soils [17]. Indeed, we found positive linear correlation between C_{org} and Br ($R^2 = 0.92$) or I ($R^2 = 0.88$) in the top soil of the study area, confirming Br and I enrichments via strong adsorption (Fig. 19). The C_{org} contents in mid- and high-elevation alpine forests and grasslands (Fig. 13, a) closely aligned spatially with Br and I enrichments, thus further suggested that Br and I mainly occur in their adsorbed form. Iodine is restricted to the medium and high elevation areas due to soil organic

matter adsorption, making it difficult to enter the piedmont basin. This is also the main reason for the local iodine deficiency in the piedmont basin [18].

Mechanisms driving the macroscopic distribution of heavy metals

Heavy metals are metal elements with a density of 4.5 g/cm^3 or more. Of the 60 naturally occurring metal elements with atomic numbers from 23 (V) to 92 (U), fifty-four have a density greater than 4.5 g/cm^3 . The following 10 metals are classified as heavy metals in industrial applications: Cu, Pb, Zn, Sn, Ni, Co, Sb, Hg, Cd and Bi. Because arsenic has a toxicity similar to that of heavy metals and a density of 5.73 g/cm^3 , it is also included as a heavy metal when it occurs in the environment. According to the characteristics of the parent materials in the study area, we selected Pb, Zn, Cd, Ag, Cu, Ni, Co, Ti and V for our investigation and discussion.

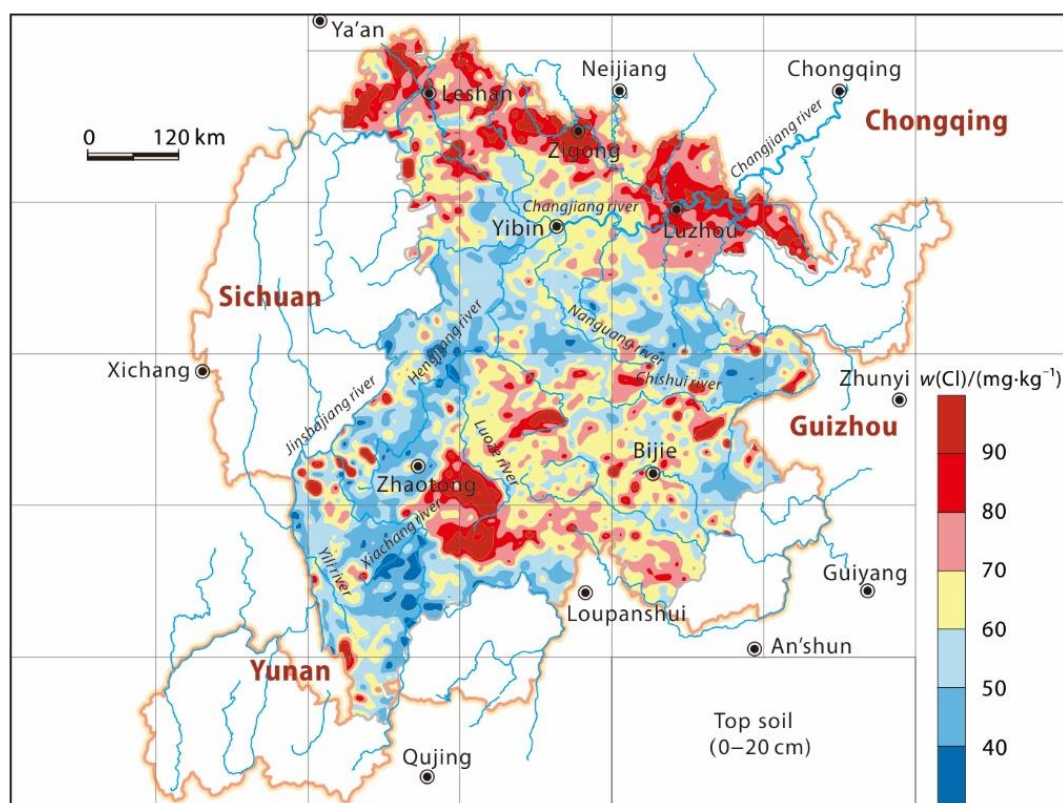


Fig. 16. Chlorine distribution pattern in the top (0–20 cm) soil of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 16. Схема распределения Cl в верхних (0–20 см) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

Lead, Zn, Cd and Ag. The study area is rich in Pb, Zn and Ag resources. Currently, more than 400 Pb-Zn deposits have been discovered, including 5 large and 14 medium-size deposits. There are 14 silver deposits (including one large and three medium-size deposits). Lead, Zn and Ag deposits are mainly distributed in the Yiliang – Hezhang, Huize – Weining and Butuo – Qiaojia mine-cluster areas. Presently, there are more than 120 Pb, Zn and Ag mines, among which 42 are in operation and the rest suspended or closed.

The distributions of Pb, Zn, Ag and Cd showed that the concentration centers of Pb (Fig. 20), Zn (Fig. 21), Ag (Fig. 22) and Cd (Fig. 23) in both top and deep soils were strictly controlled by the distributions of Pb, Zn and Ag deposits in the Pb-Zn-Ag mine clusters. Comparison of Pb, Zn, Ag and Cd levels in both soils revealed clear cumulative distribution patterns for all elements regardless of their absolute contents. Additionally, regional Pb, Zn, Ag and Cd peaks occurred in the top soils of the Weining – Hezhang area in Guizhou Province.

Our analyses showed that the cumulation patterns of Pb, Zn, Ag and Cd in the top soils changed with elevation. Lead, Zn and Cd contents peaked at the same elevation as the Pb, Zn and Ag deposits (Fig. 24, a–c), with decreasing Ag content as elevation declines (Fig. 24, d). Compared to deep soil, concentrations of Pb, Zn, Ag and Cd in the top soil were higher by 29.8 %, 6.8 %, 18.9 % and 169 %, respectively. The difference was more than three orders of magnitude. For Cd, its contents in the top soil were 142 %, 235 % and 102 % higher than in the deep soil at elevations below 1,800 m, 1,800–2,700 m and above 2,700 m, respectively. For Zn, meanwhile, 2,400 m was the point of division: at below 2,400 m, the Zn content in the top soil was 6–18 % higher than in the deep soil, and the difference grew as elevation decreases. At above 2,400 m, the differences were further reduced and even became negative at above 2,800 m. The changes in Pb and Ag concentrations in the top soil, however, were significantly different from those for Zn and Cd. As elevation decreases, Pb and Ag contents in the top soil became increasingly similar to that in the deep soil.

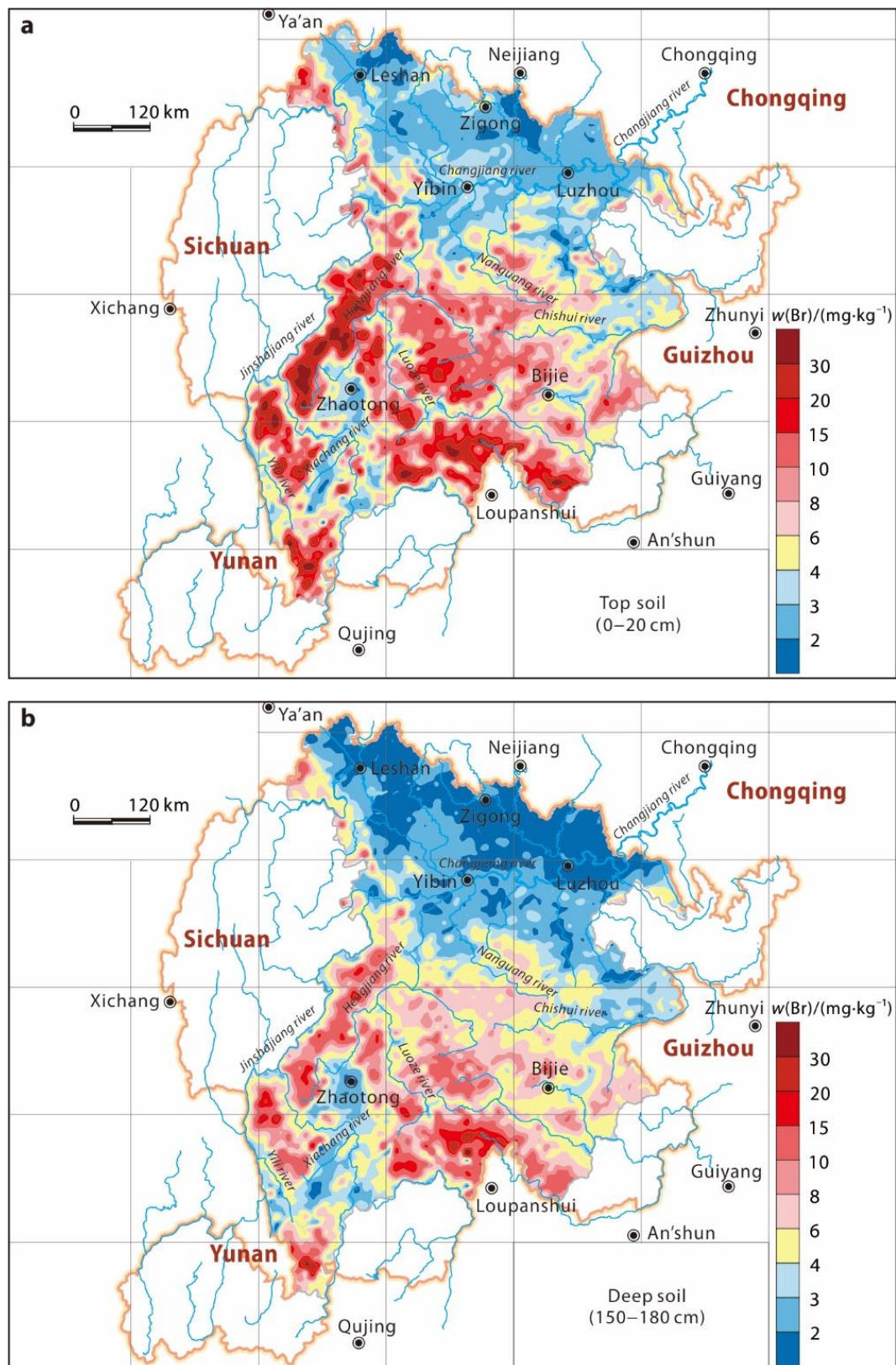


Fig. 17. Bromine distribution pattern in the top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area

Рис. 17. Схема распределения Br в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

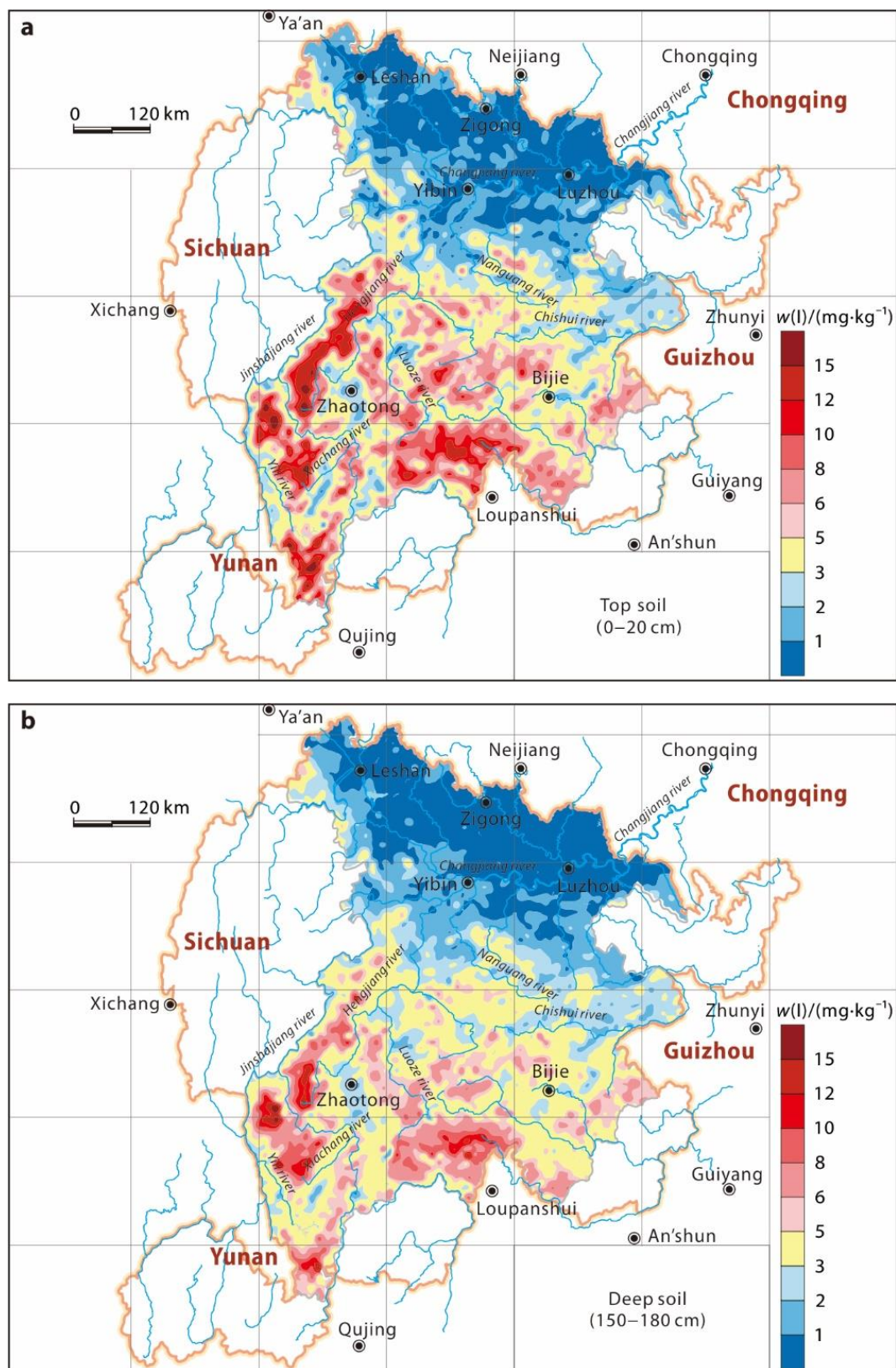


Fig. 18. Iodine distribution pattern in the top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 18. Схема распределения I в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

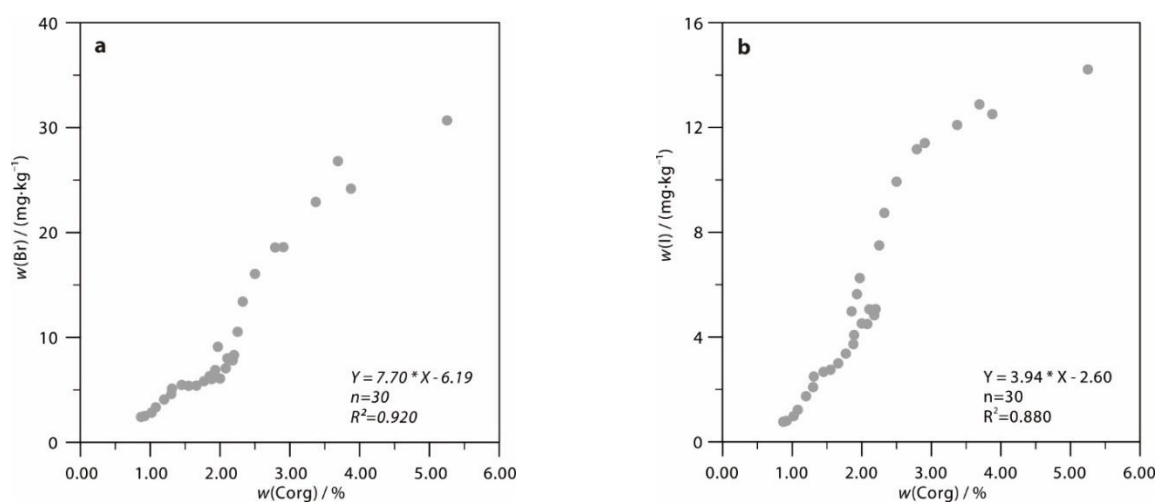


Fig. 19. Correlation between C_{org} and Br (a) or I (b) in the top soil of the Zhaotong – Bijie – Yibin – Quzhou – Leshan area

Рис. 19. Корреляция между C_{org} и Br (a) или I (b) в верхних почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

In certain geographical areas, the epigenetic geochemical behavior of elements are mainly depended on their geochemical properties and organic matters, pH and clay minerals in the top soil. A partial analysis of correlations between Pd, Zn, Ag or Cd levels in soils and organic matter or pH indicated that, the main driver of the geochemical behavior of Ag and Pb in an epigenetic environment is organic carbon, while that for Zn and Cd is pH (Table 3). Because Ag can be strongly adsorbed and fixed by organic matter in epigenetic environments, while Zn has relatively high solubility in acidic environments, Ag enrichment (Figs. 22, a and 24, d) and Zn depletion (Figs. 21, a and 24, b) were noted in the top soils of alpine meadows and forests.

The magnitude of increasing Pb and Cd contents in the top and deep soils at the elevation of the deposits was significantly larger than that in other areas (Fig. 24, a,c). Also, we observed Cd, Pb, Zn and Ag pollution in some regions. It suggests extreme levels of human disturbance, including mining and metal extraction, at the elevation of the deposits. A detailed literature review suggested that the Hezhang – Weining region was once (over 300 years ago) a center for indigenous zinc smelting activities, in addition to the large-, medium-, and small-size Pb and Zn mines such as Tianqiao (Zhugongtang), Wuliping, Shaojiwan, Yadu and Zhushachang mines that are currently in operation or closed. The slag formed during mining has been spread throughout the region [19, 20] and is the main source of

Cd, Pb, Zn and Ag pollution (Fig. 25). In a survey of Cd contents in corn in the above-mentioned areas found that 12.1 % of the investigated corn had Pb and Cd contents higher than the allowable amounts [21]. Such finding is rare in corn-growing areas nationwide.

Copper, Ni, Co, Ti and V. The Emeishan LIP is generally considered to be the product of a mantle-plume event [22, 23] and characterized by high Cu, Ni, Co, Cr, Ti and V contents. In the southwestern part of the study region, areas with high Cu, Ni, Co, Cr, Ti and V contents in the top and deep soils overlap with the Emeishan basalt area, and the same holds true for the carbonate area in the southeastern part of the study region. This indicates that the spatial distributions of Cu, Ni, Co, Cr, Ti and V are mainly controlled by the Emeishan basalt and carbonate rock parent materials (Figs. 26–31). We showed in a statistical analysis that the average relative change rates of Cu, Ni, Co, Cr, Ti and V contents in the top soil, with respect to that in the deep soil (defined as (content in the top soil – content in the deep soil) / content in the deep soil) × 100) were -5.9 %, -5.31 %, -6.09 %, -6.52 %, -4.8 % and -2.58 %, respectively, demonstrating that the concentrations of these elements were all slightly lower in the top soil. In a correlation analysis comparing elevation with elemental content, we showed that the rates of Co and Ni loss in the top soil increased with increasing elevation (Fig. 32, a,b). The average rates of change of Cu, Cr, Ti and V contents in the top soil at different

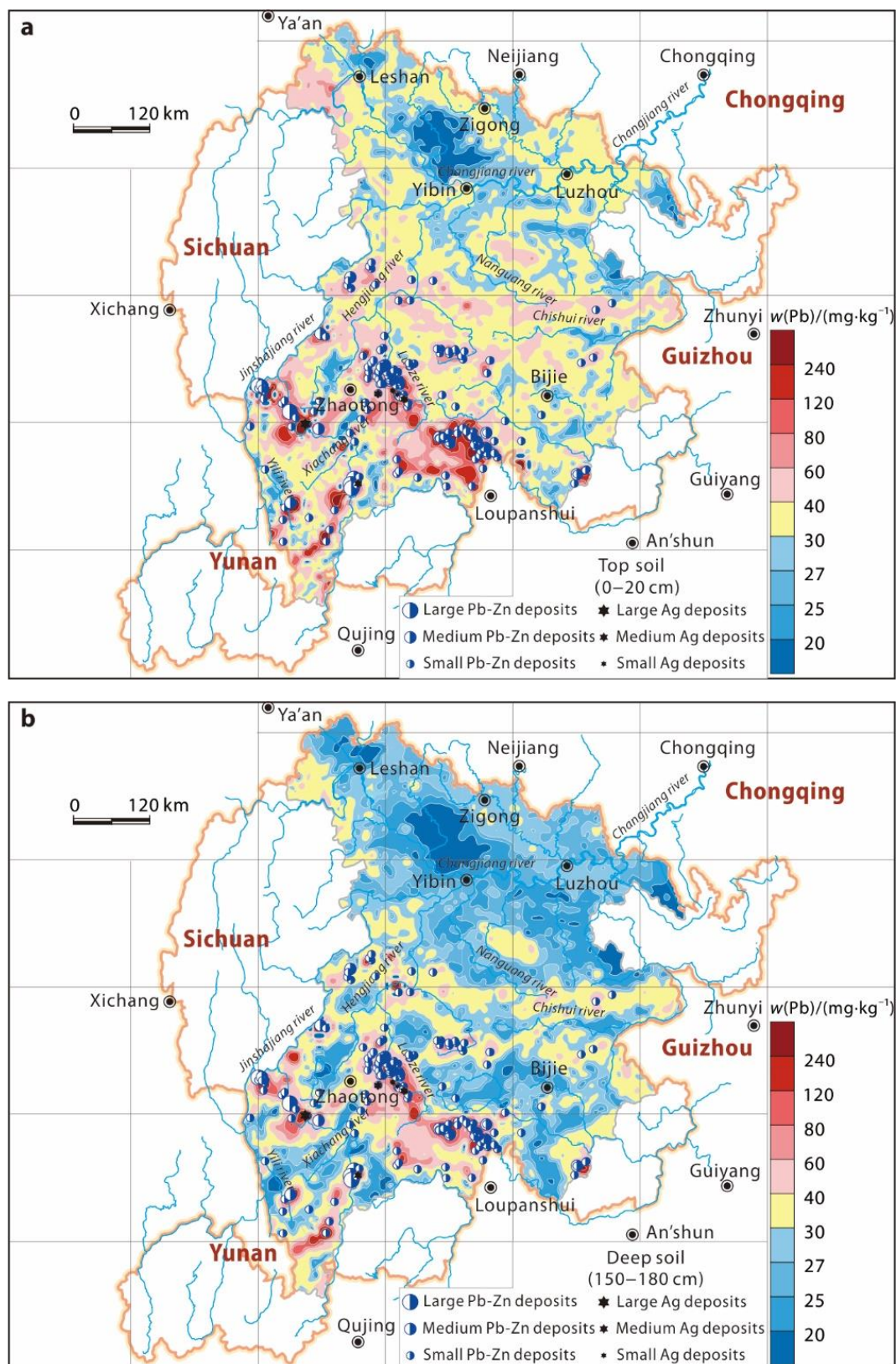


Fig. 20. Lead distribution pattern in the top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 20. Схема распределения Pb в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

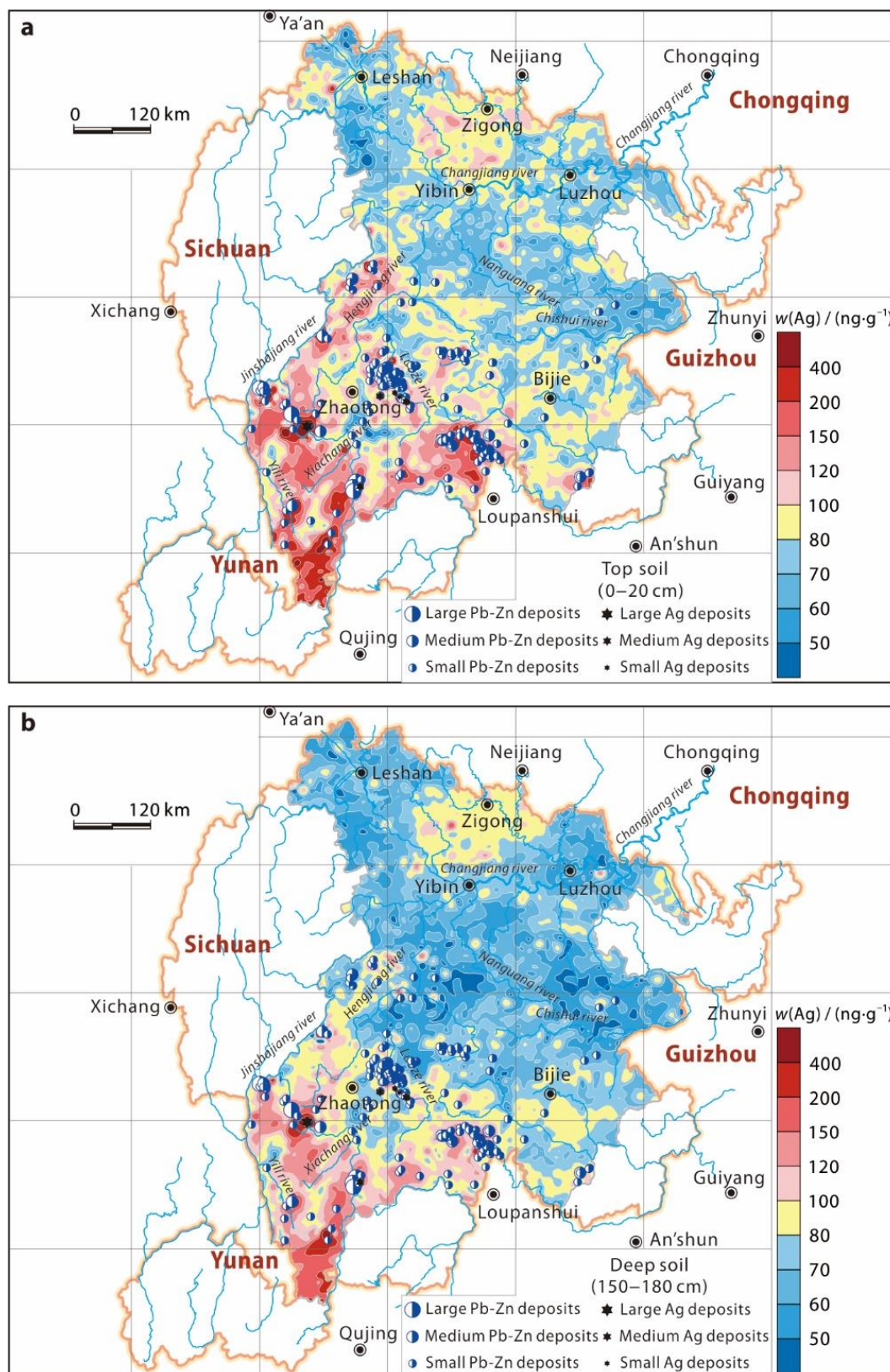


Fig. 22. Silver distribution pattern in the top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 22. Схема распределения Ag в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

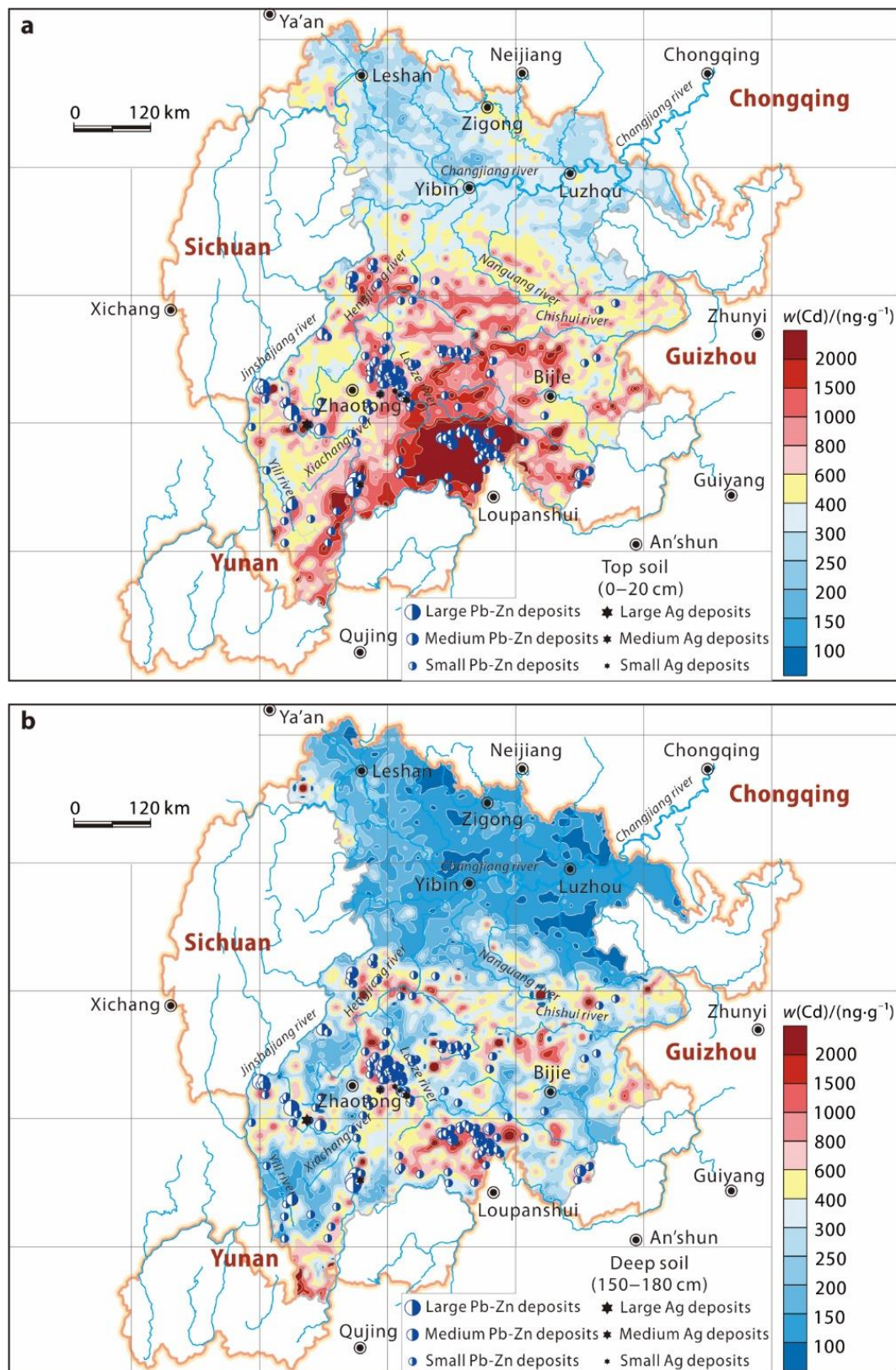


Fig. 23. Cadmium distribution pattern in the top (0–20 cm) (a) and deep (150–180 cm) (b) soil of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 23. Схема распределения Cd в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань



elevations were relatively consistent (Fig. 32, c–f), indicating the change of top soil contents of

these five elements is essentially controlled by the same factors throughout the region.

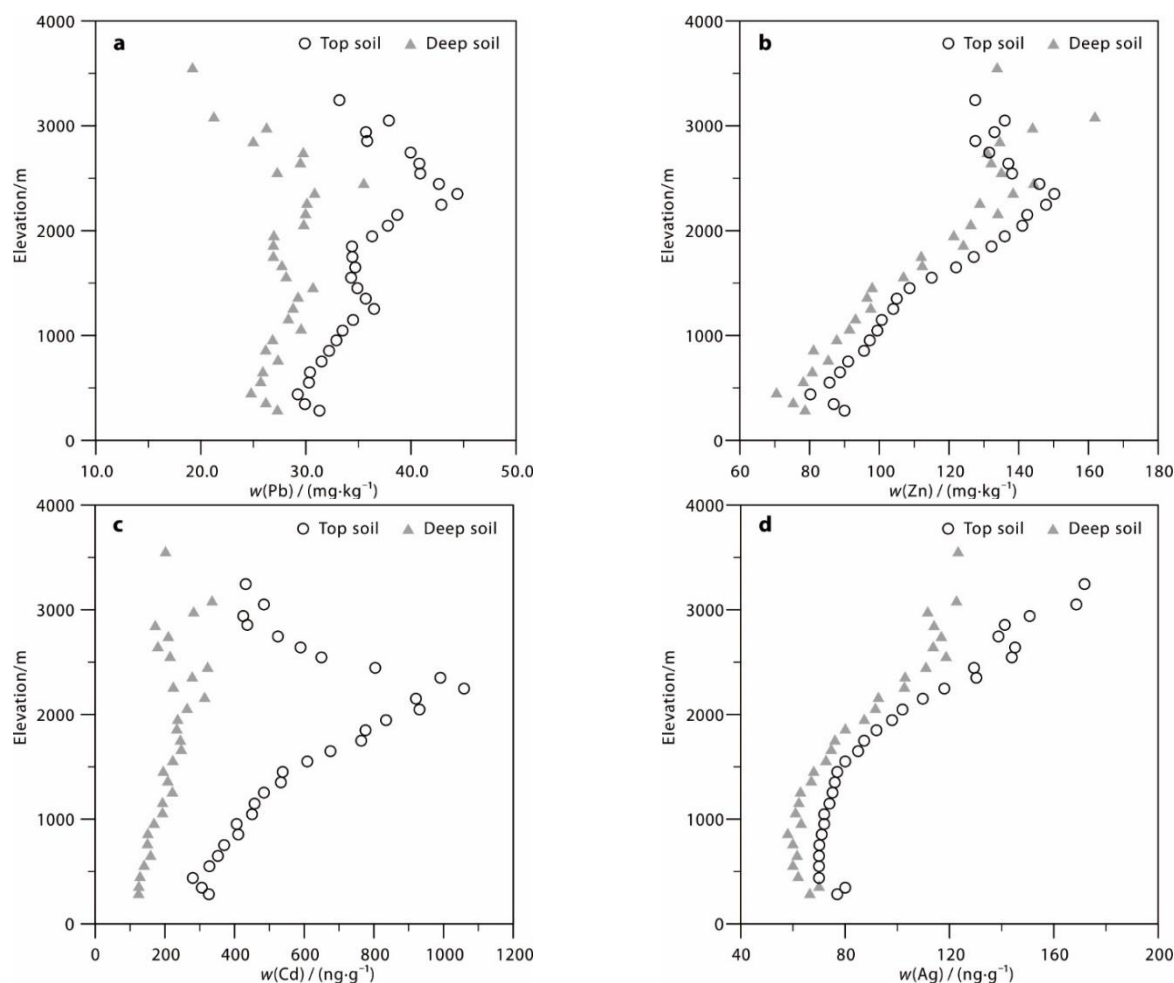


Fig. 24. Plot of Pb (a), Zn (b), Cd (c) and Ag (d) contents in the top and deep soils vs. elevation in the Zhaotong – Bijie – Yibin – Luzhou – Leshan area

Рис. 24. График содержания Pb (a), Zn (b), Cd (c) и Ag (d) в верхних и глубоких почвах в зависимости от глубины в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

Table 3. Partial correlation analysis of Pb, Zn, Ag and Cd

with respect to C_{org} and pH in soils of the Zhaotong – Bijie – Yibin – Leshan – Luzhou area

Таблица 3. Частичный корреляционный анализ содержаний Pb, Zn, Ag и Cd

в соответствии с содержанием C_{org} и pH в почвах района Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

Element	Test parameters	pH (constant C_{org})	C_{org} (constant pH)
Ag	Correlation	0.055	0.817
	Significance (two-tailed)	0.784	0
	df	25	25
Cd	Correlation	-0.593	-0.323
	Significance (two-tailed)	0.001	0.101
	df	25	25
Pb	Correlation	-0.08	0.655
	Significance (two-tailed)	0.692	0
	df	25	25
Zn	Correlation	-0.583	-0.257
	Significance (two-tailed)	0.001	0.195
	df	25	25

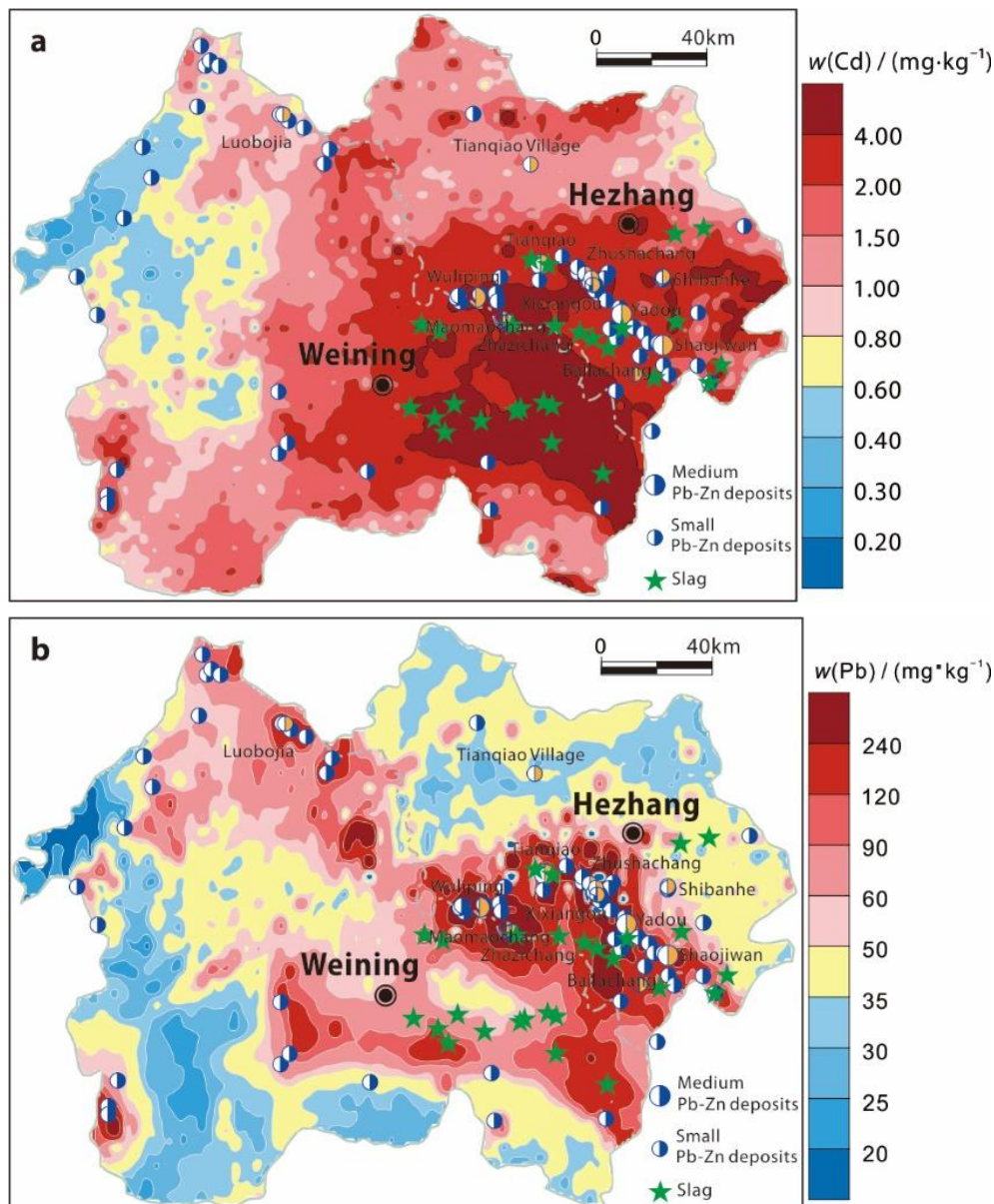


Fig. 25. Geochemical map of the Weining – Hezhang area, showing the locations of Zn smelting slag (adapted from [19]) and Cd (a) and Pb (b) contents in the top soils
Рис. 25. Геохимическая карта района Вейнинг – Хечжан с указанием местонахождения цинкового шлака (по данным источника [19]), Cd (a) и Pb (b), содержащихся в верхних почвах

The analysis of driving factors of elemental content (Table 4) demonstrated that, the partial correlation coefficients of Cu, Ni, Co, Cr, Ti and V with respect to pH, were greater than those to organic carbon. This suggests that the loss of these elements in epigenetic environments is mainly driven by pH.

Conclusions and implications

Our study on driving mechanisms of the macroscopic distribution of elements in the top and deep soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan region clearly showed the

following: the geological environment determines the natural concentrations of chemical elements in soil; epigenetic geochemical processes re-shape the microscopic distribution of elements in the top soil; biogeochemical processes drive the evolution of land quality; intensive human activities (such as mining) have disruptive impact on the natural evolution of elemental distribution. A better understanding of the dynamic geochemical processes on the Earth's surface can help to develop a theoretical framework, within which we can study the regional ecology formed through the interactions of topography, hydrology

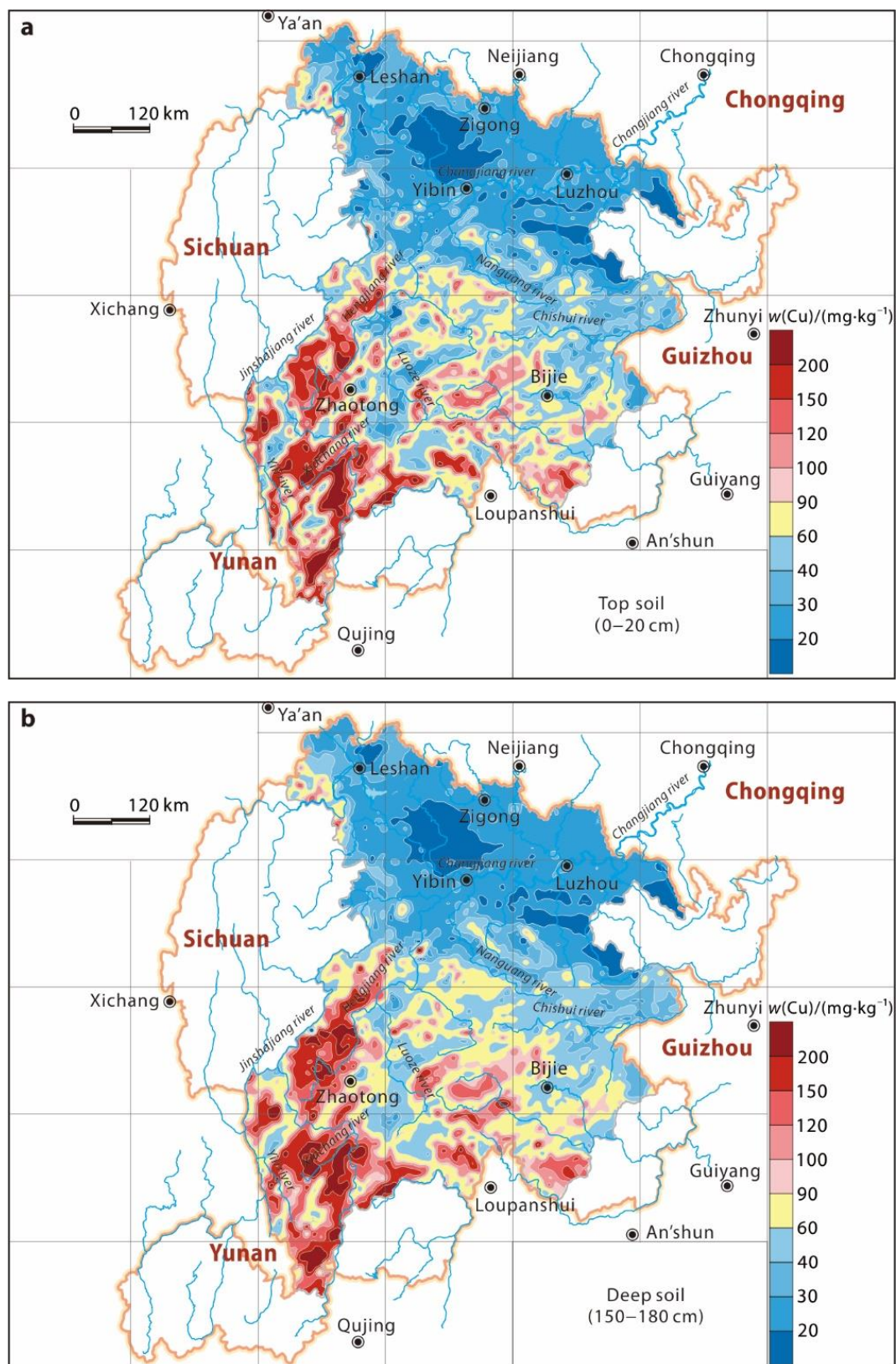


Fig. 26. Copper distribution pattern in the top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 26. Схема распределения Си в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

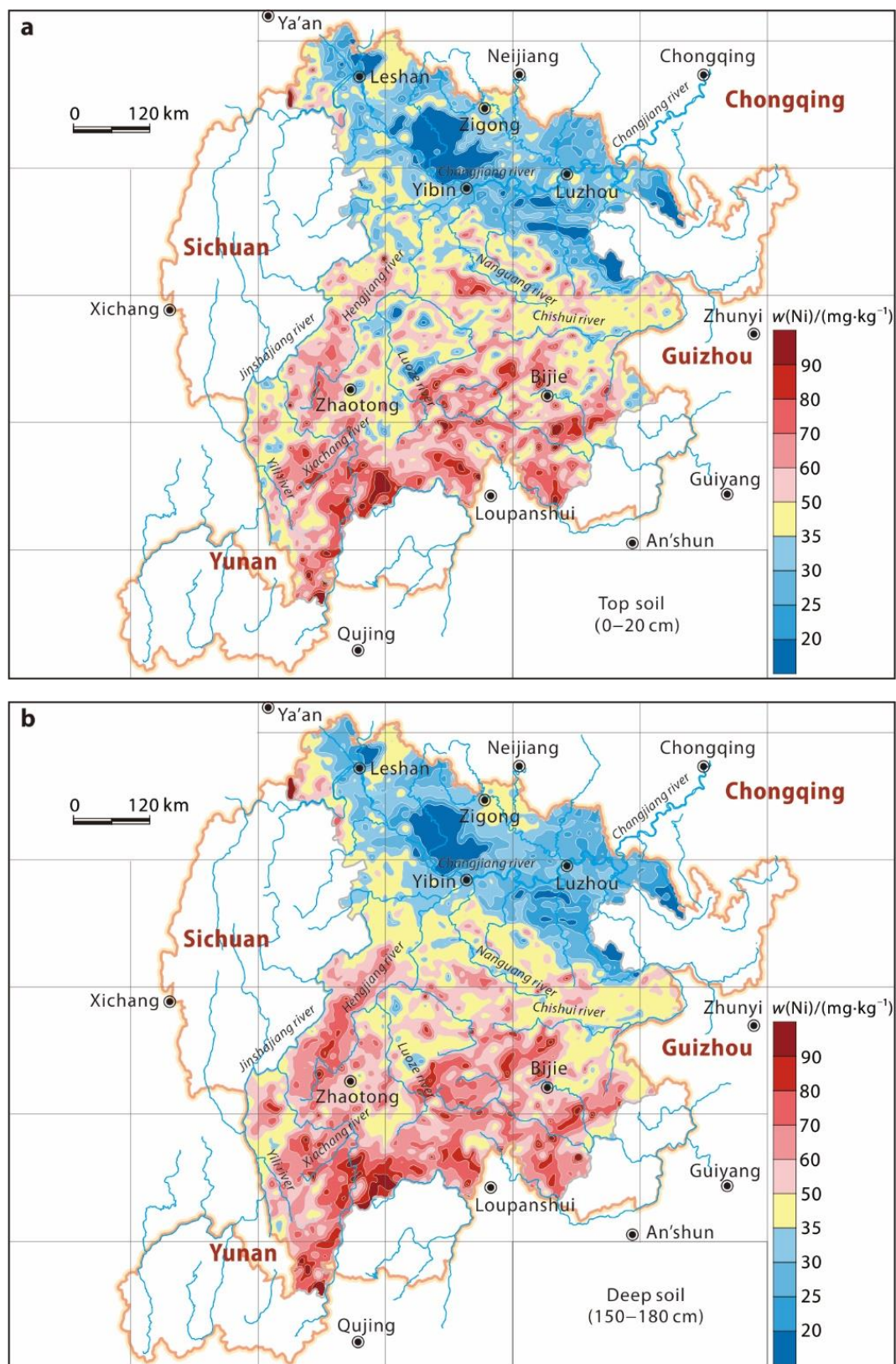


Fig. 27. Nickel distribution pattern in the top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 27. Схема распределения Ni в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

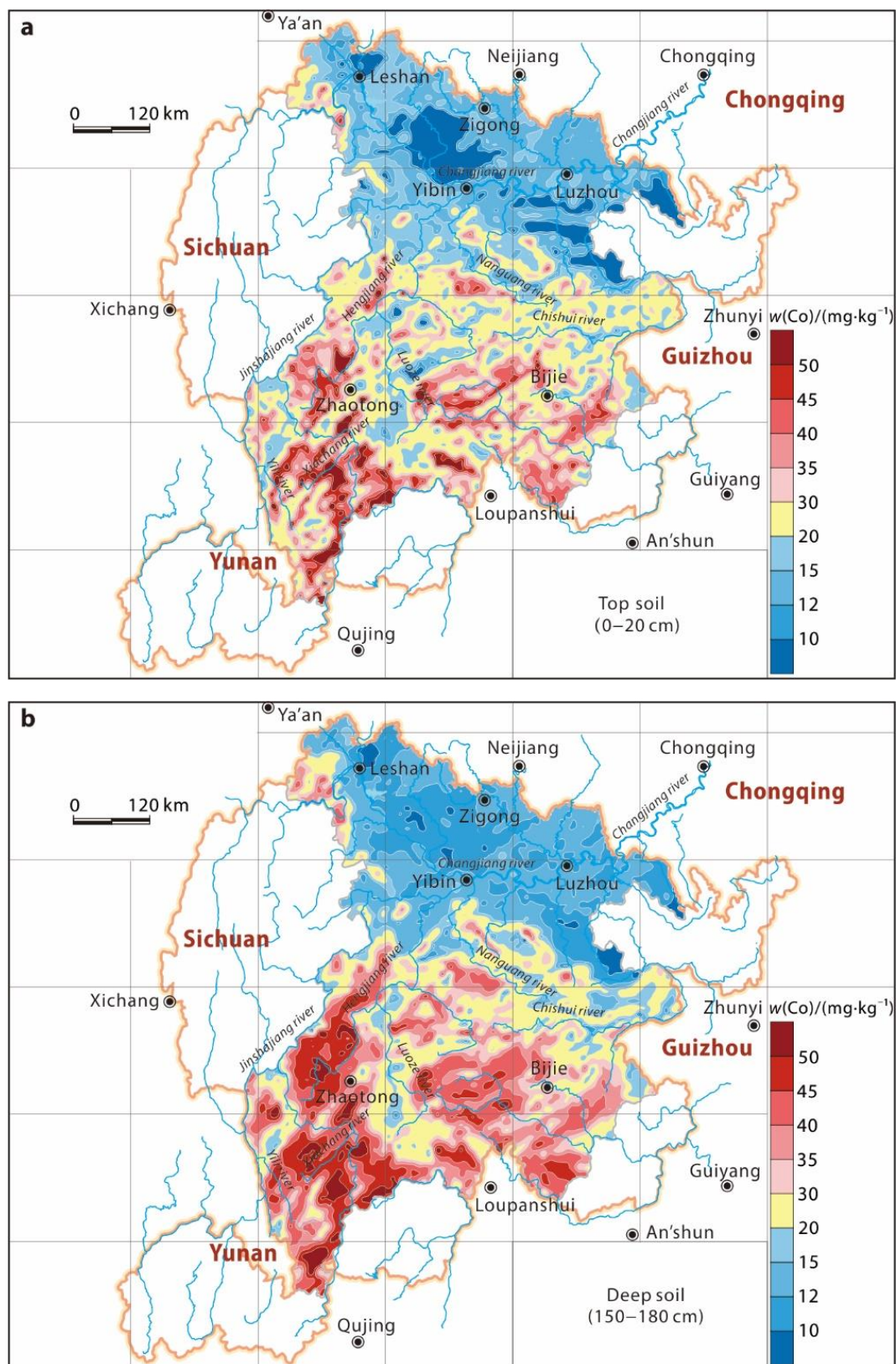


Fig. 28. Cobalt distribution pattern in the top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 28. Схема распределения Co в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

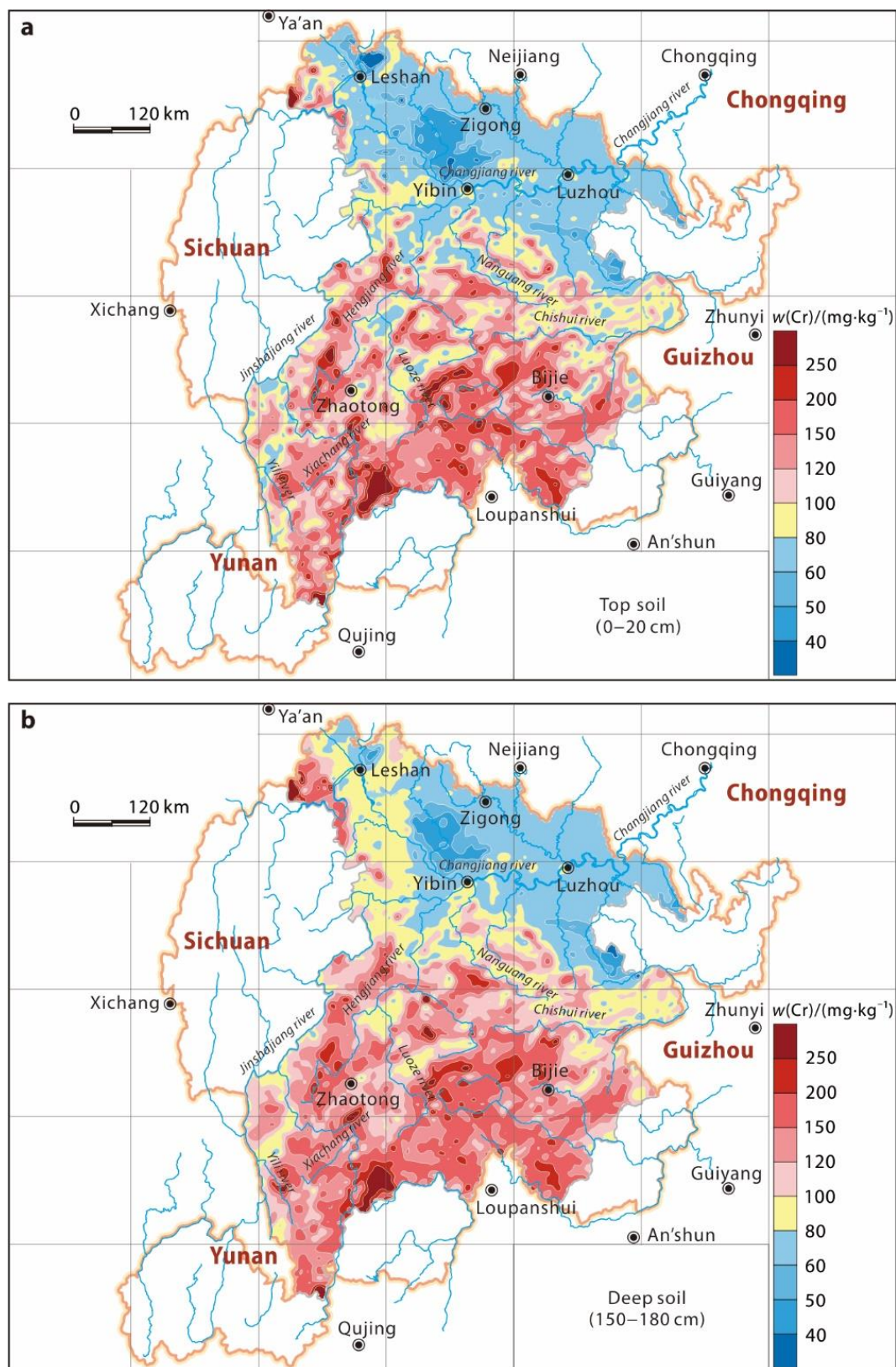


Fig. 29. Chromium distribution pattern in the top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 29. Схема распределения Cr в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

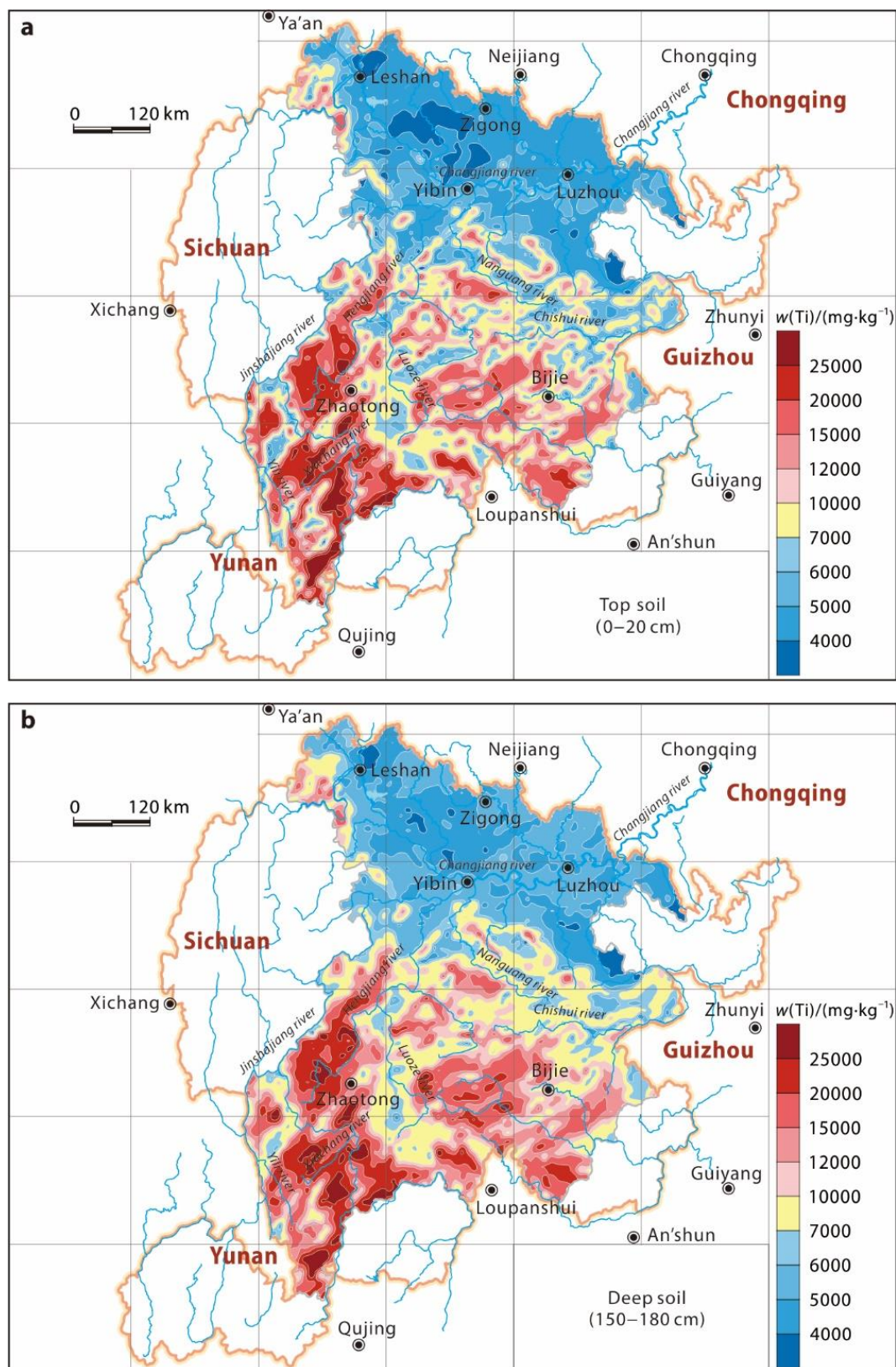


Fig. 30. Titanium distribution pattern in the top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 30. Схема распределения Ti в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

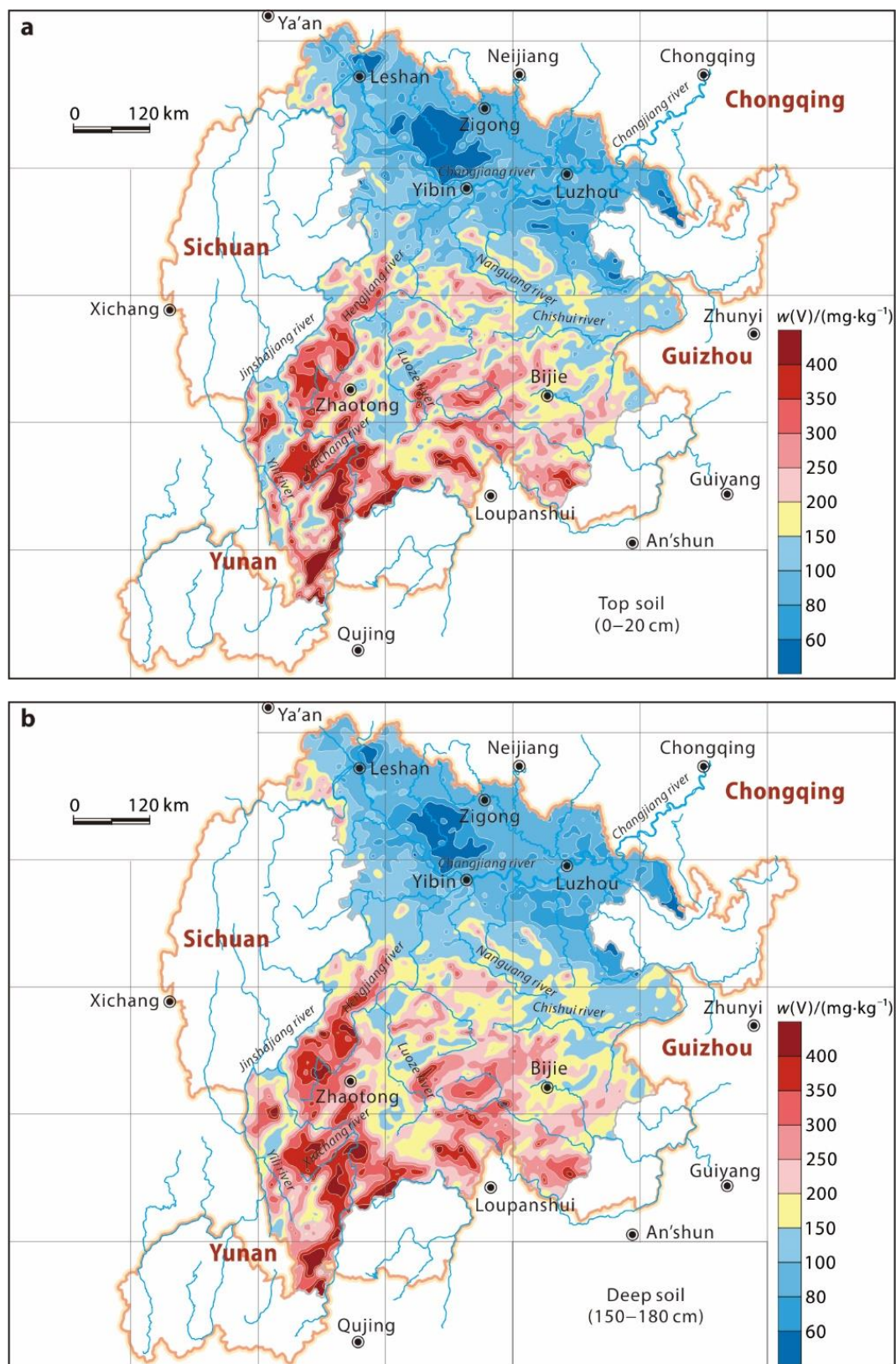


Fig. 31. Vanadium distribution pattern in the top (0–20 cm) (a) and deep (150–180 cm) (b) soils of the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Рис. 31. Схема распределения V в верхних (0–20 см) (a) и глубоких (150–180 см) (b) почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

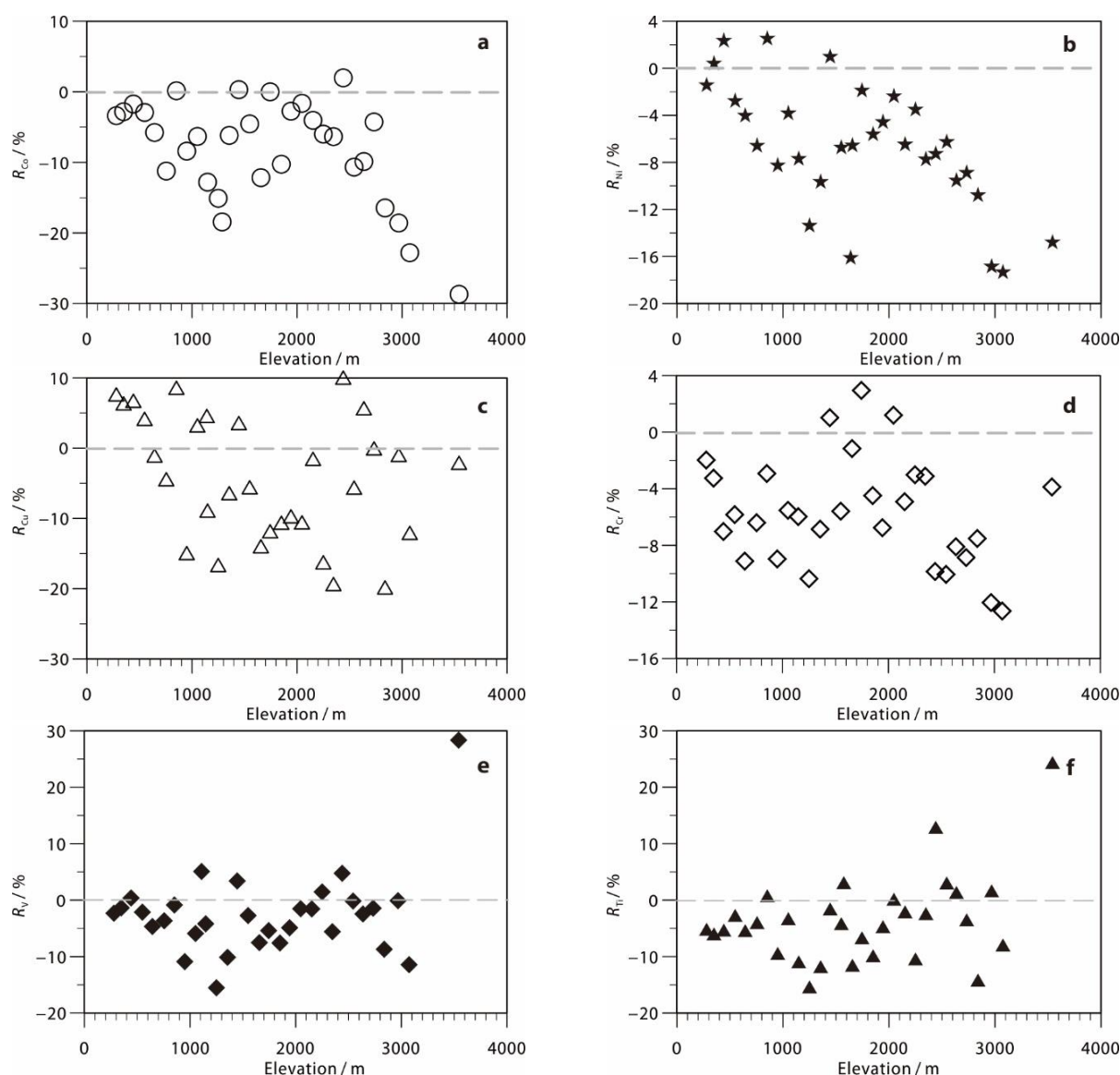


Fig. 32. Relative change rate (R_i) of Co (a), Ni (b), Cu (c), Cr (d), V (e) and Ti (f) contents in the top soil compared to that in the deep soil at different elevations in the Zhaotong – Bijie – Yibin – Luzhou – Leshan area

$$R_i = ((\text{content in the top soil} - \text{content in the deep soil}) / \text{content in the deep soil}) \times 100$$

Рис. 32. Относительная скорость изменения (R_i) содержания Co (a), Ni (b), Cu (c), Cr (d), V (e) и Ti (f) в верхних почвах по сравнению с глубокими почвами на разных высотах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

$$R_i = ((\text{содержание элемента в верхних почвах} - \text{содержание элемента в глубоких почвах}) / \text{содержание элемента в глубоких почвах}) \times 100$$

and land cover. Such framework has the potential for comprehensively integrating different scientific theories of Earth's systems. To study the physical, chemical and biodynamic processes on the Earth's surface, a new comprehensive survey and monitoring system for mountains, water bodies, forests, farmlands and grasslands is required. This system could help us determine the current natural resource usage, land quality, ecological quality and natural evolution patterns, and

assess land quality and future trends, so as to provide guidance for spatial planning and management nationwide.

China is a vast country with complex and diverse geographical landscapes [24]. In this study, we only analyzed the southwestern area characterized by mountains and gorges, and primarily revealed the mechanisms driving the macroscopic distribution of elements in the regional soils. Similar studies should be conducted in



Table 4. Results of partial correlation analysis of Cu, Ni, Co, Ti and V with respect to pH and C_{org} in soil in the Zhaotong – Bijie – Yibin – Luzhou – Leshan area
Таблица 4. Результаты частичного корреляционного анализа Cu, Ni, Co, Ti и V по отношению к pH и C_{org} в почвах в районе Чжаотун – Бицзе – Ибинь – Лучжоу – Лэшань

Element	Test parameter	pH (constant C _{org})	C _{org} (constant pH)
Cu	Correlation	-0.841	0.407
	Significance (two-tailed)	0	0.035
	df	25	25
Ni	Correlation	-0.588	-0.337
	Significance (two-tailed)	0.001	0.086
	df	25	25
Co	Correlation	-0.741	-0.504
	Significance (two-tailed)	0	0.007
	df	25	25
Cr	Correlation	-0.58	-0.303
	Significance (two-tailed)	0.002	0.124
	df	25	25
Ti	Correlation	-0.847	0.403
	Significance (two-tailed)	0	0.037
	df	25	25
V	Correlation	-0.769	0.272
	Significance (two-tailed)	0	0.169
	df	25	25

future to analyze a variety of landscapes, including forests and swampy landscapes, humid and semi-humid low mountains, semi-arid hilly landscapes, alpine lakes in hilly landscapes, arid and semi-arid or humid and semi-humid alpine mountains, arid Gobi relict mountains, karst areas, alluvial plains, and colluvial deposits in the Gobi desert. Such studies would represent considerable advances in epigenetic geochemical dynamics and help us to establish a solid foundation for the development of scientific theories of the Earth's systems [25, 26].

Currently, surveys on the quantity, quality and ecological status of various natural resources in China are disconnected rather than combined in a single system. Presently, the Ministry of Natural Resources is responsible for all natural resource assets. It formulates land use regulations and

conducts ecological protection and restoration. However, a comprehensive survey and monitoring system, integrating the quantity, quality and ecological conditions of all natural resources based on their various transport patterns, remains to be developed. Moreover, chemical elements are the basic components of natural resources. For example, our findings on the epigenetic geochemical dynamic processes and mutual interactions of resources components, in the southwestern mountain and gorge regions, suggest that changes in chemical elemental contents in the environment are extremely sensitive indicators of environmental changes. Hence, we recommend that the chemical compositions of natural resources should also be included in the procedures during the development of future survey and monitoring systems.

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Hangxin Cheng, Min Peng, Chuandong Zhao, Wei Han, Huiyan Wang, Qiaolin Wang, Fan Yang, Fugui Zhang, Chengwen Wang, Fei Liu, Yalong Zhou, Shiqi Tang, Kuo Li, Ke Yang, Zheng Yang, Xiaomeng Cheng, Ziwan Chen, Hua Zhang, Chunhu Mo are the authors of the article, hold equal copyright and bear equal responsibility for plagiarism.

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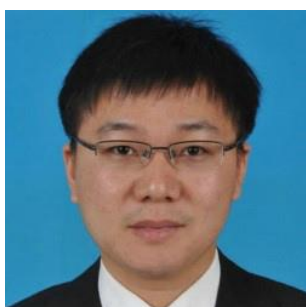
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Information about the authors / Сведения об авторах



Hangxin Cheng,
Doctor's Degree,
Professor,
Institute of Geophysical & Geochemical Exploration,
Chinese Academy of Geological Sciences,
84 Jinguang Road, Langfang 065000, China,
✉ e-mail: 916679036@qq.com
Чэн Хансинь,
доктор,
профессор,
Институт геофизических и геохимических исследований
Китайской академии геологических наук,
065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,
✉ e-mail: 916679036@qq.com



Min Peng,
Master's Degree,
Engineer,
Institute of Geophysical & Geochemical Exploration,
Chinese Academy of Geological Sciences,
84 Jinguang Road, Langfang 065000, China,
School of Geosciences and Resources,
China University of Geosciences (Beijing),
29 Xueyuan Road, Beijing 100083, China,
e-mail: pm-ant@qq.com
Пэн Минь,
магистр,
инженер,
Институт геофизических и геохимических исследований
Китайской академии геологических наук,
065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,
Школа геологических наук и природных ресурсов,
Китайский геологический университет (Пекин),
100083, г. Пекин, ул. Сюеюань Роуд, 29, Китай,
e-mail: pm-ant@qq.com



Chuandong Zhao,
Doctor's Degree,
Professor,
Institute of Geophysical & Geochemical Exploration,
Chinese Academy of Geological Sciences,
84 Jinguang Road, Langfang 065000, China,
e-mail: Zhaochuandong@igge.cn
Чжао Чуандун,
доктор,
профессор,
Институт геофизических и геохимических исследований
Китайской академии геологических наук,
065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,
e-mail: Zhaochuandong@igge.cn

Wei Han,
Master's Degree,
Engineer,
Institute of Geophysical & Geochemical Exploration, Chinese Academy of Geological Sciences,
84 Jinguang Road, Langfang 065000, China,
e-mail: 331224192@qq.com

**Хань Вэй,**

магистр,
инженер,

Институт геофизических и геохимических исследований Китайской академии геологических наук,
065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,
e-mail: 331224192@qq.com

Huiyan Wang,

Master's Degree,
Engineer,

Institute of Geophysical & Geochemical Exploration, Chinese Academy of Geological Sciences,
84 Jinguang Road, Langfang 065000, China,
e-mail: wanghuiyan@igge.cn

Ван Хуйянь,

магистр,
инженер,

Институт геофизических и геохимических исследований Китайской академии геологических наук,
065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,
e-mail: wanghuiyan@igge.cn

Qiaolin Wang,

Master's Degree,
Engineer,

Institute of Geophysical & Geochemical Exploration, Chinese Academy of Geological Sciences,
84 Jinguang Road, Langfang 065000, China,
e-mail: 408409647@qq.com

Ван Цяолин,

магистр,
инженер,

Институт геофизических и геохимических исследований Китайской академии геологических наук,
065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,
e-mail: 408409647@qq.com

Fan Yang,

Doctor's Degree,
Senior Engineer,

Institute of Geophysical & Geochemical Exploration, Chinese Academy of Geological Sciences,
84 Jinguang Road, Langfang 065000, China,
e-mail: yangfan@igge.cn

Ян Фань,

доктор,
старший инженер,

Институт геофизических и геохимических исследований Китайской академии геологических наук,
065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,
e-mail: yangfan@igge.cn

Fugui Zhang,

Master's Degree,
Senior Engineer,

Institute of Geophysical & Geochemical Exploration, Chinese Academy of Geological Sciences,
84 Jinguang Road, Langfang 065000, China,
e-mail: zhangfugui@igge.cn

Чжан Фугуй,

магистр,
старший инженер,

Институт геофизических и геохимических исследований Китайской академии геологических наук,
065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,
e-mail: zhangfugui@igge.cn



Chengwen Wang,

Master's Degree,

Engineer,

Institute of Geophysical & Geochemical Exploration, Chinese Academy of Geological Sciences,

84 Jinguang Road, Langfang 065000, China,

e-mail: 187311220@qq.com

Ван Чэнвэнь,

магистр,

инженер,

Институт геофизических и геохимических исследований Китайской академии геологических наук,

065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,

e-mail: 187311220@qq.com

Fei Liu,

Doctor's Degree,

Senior Engineer,

Institute of Geophysical & Geochemical Exploration, Chinese Academy of Geological Sciences,

84 Jinguang Road, Langfang 065000, China,

e-mail: 29619105@qq.com

Лю Фэй,

доктор,

старший инженер,

Институт геофизических и геохимических исследований Китайской академии геологических наук,

065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,

e-mail: 29619105@qq.com

Yalong Zhou,

Master's Degree,

Senior Engineer,

Institute of Geophysical & Geochemical Exploration, Chinese Academy of Geological Sciences,

84 Jinguang Road, Langfang 065000, China,

e-mail: zhouyalong@igge.cn

Чжоу Ялун,

магистр,

старший инженер,

Институт геофизических и геохимических исследований Китайской академии геологических наук,

065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,

e-mail: zhouyalong@igge.cn

Shiqi Tang,

Master's Degree,

Assistant Engineer,

Institute of Geophysical & Geochemical Exploration, Chinese Academy of Geological Sciences,

84 Jinguang Road, Langfang 065000, China,

e-mail: 642191779@qq.com

Тан Шици,

магистр,

ассистент инженера,

Институт геофизических и геохимических исследований Китайской академии геологических наук,

065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,

e-mail: 642191779@qq.com

Kuo Li,

Master's Degree,

Engineer,

Institute of Geophysical & Geochemical Exploration, Chinese Academy of Geological Sciences,

84 Jinguang Road, Langfang 065000, China,

e-mail: likuo@igge.cn

**Ли Ко,**

магистр,
инженер,

Институт геофизических и геохимических исследований Китайской академии геологических наук,
065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,
e-mail: likuo@igge.cn

Ke Yang,

Doctor's Degree,
Senior Engineer,

Institute of Geophysical & Geochemical Exploration, Chinese Academy of Geological Sciences,
84 Jinguang Road, Langfang 065000, China,
e-mail: 47384668@qq.com

Ян Кэ,

доктор,
старший инженер,

Институт геофизических и геохимических исследований Китайской академии геологических наук,
065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,
e-mail: 47384668@qq.com

Zheng Yang,

Master's Degree,
Assistant Engineer,

Institute of Geophysical & Geochemical Exploration, Chinese Academy of Geological Sciences,
84 Jinguang Road, Langfang 065000, China,
e-mail: yangzheng@igge.cn

Ян Чжэн,

магистр,
ассистент инженера,

Институт геофизических и геохимических исследований Китайской академии геологических наук,
065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,
e-mail: yangzheng@igge.cn

Xiaomeng Cheng,

Master's Degree,
Assistant Engineer,

Institute of Geophysical & Geochemical Exploration, Chinese Academy of Geological Sciences,
84 Jinguang Road, Langfang 065000, China,
e-mail: 309498905@qq.com

Чэн Сяомэн,

магистр,
ассистент инженера,

Институт геофизических и геохимических исследований Китайской академии геологических наук,
065000, г. Ланфан, ул. Цзиньгуан Роуд, 84, Китай,
e-mail: 309498905@qq.com

Ziwan Chen,

Master's Degree,
Senior Engineer,

Yunnan Institute of Geological Survey,
84 Renmin East Road, Kunming 650216, China,
e-mail: ChenZW_cdut@outlook.com

Чэнь Цзывань,

магистр,
старший инженер,

Юньнаньский институт Геологической службы,
650216, г. Куньмин, ул. Жэньминь Ист Роуд, 84, Китай,
e-mail: ChenZW_cdut@outlook.com



Hua Zhang,

Bachelor's Degree,
Senior Engineer,
Sichuan Geological Survey,
25 Renming Road, Chengdu 610081, China,
e-mail: 258155276@qq.com

Чжан Хуа,

бакалавр,
старший инженер,
Сычуаньская геологическая служба,
610081, г. Чэнду, ул. Жэньминь Ист Роуд, 25, Китай,
e-mail: 258155276@qq.com

Chunhu Mo,

Bachelor's Degree,
Senior Engineer,
Guizhou Geological Survey,
171 Shilin West Road, Guiyang 550081, China,
e-mail: 196189077@qq.com

Мо Чуньху,

бакалавр,
старший инженер,
Геологическая служба Гуйчжоу,
550081, г. Гуйян, ул. Шилинь Вест Роуд, 171, Китай,
e-mail: 196189077@qq.com