

Article

National-Scale Geochemical Baseline of 69 Elements in Laos Stream Sediments

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Abstract: Geochemical baselines are crucial to explore mineral resources and monitor environmental changes. This study presents the first Laos geochemical baseline values of 69 elements. The National-scale Geochemical Mapping Project of Lao People's Democratic Republic conducted comprehensive stream sediment sampling across Laos, yielding 2079 samples collected at 1 sample/100 km², and 69 elements were analyzed. Based on the results of LGB value, R-mode factor analysis, and scatter plot analysis, this paper analyzes the relationship between the 69 elements and the geological background, mineralization, hypogene processes and human activities in the study area. The median values of element contents related to the average crustal values were: As, B, Br, Cs, Hf, Li, N, Pb, Sb, Zr, and SiO₂, >1.3 times; Ba, Be, Cl, Co, Cr, Cu, F, Ga, Mn, Mo, Ni, S, Sc, Sr, Ti, Tl, V, Zn, Eu, Al₂O₃, Tot.Fe₂O₃, MgO, CaO, and Na₂O, <0.7 times; and Ag, Au, Bi, Cd, Ge, Hg, I, In, Nb, P, Rb, Se, Sn, Ta, Th, U, W, Y, La, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and K₂O, 0.7–1.3 times. R-mode factor analysis based on principal component analysis and varimax rotation showed that they fall into 12 factors related to bedrock, (rare earth, ferrum-group, and major Al₂O₃ and K₂O elements; mineralization–Au, Sb, and As) and farming activities–N, Br, S, and C). This study provides basic geochemical data for many fields, including basic geology, mineral exploration, environmental protection and agricultural production in Laos.

Keywords: Laos; national scale; geochemical baseline; 69 elements; stream sediment



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1. Introduction

Geochemical mapping has proven to be a cost-effective exploration method for mineral resources [1–8]. Geochemists from all over the world have carried out a series of studies on sample collection, analysis and testing, map making and other means of geochemical mapping, and achieved essential results. The geochemical mapping method is the most effective method for the rapid exploration and evaluation of mineral resources [1–9]. In China, national geochemical mapping has been carried out for more than 40 years [6,7,10,11]. Since 1978, more than 2000 ore deposits have been identified by follow-up exploration within targets delineated by China's National Geochemical Mapping Project [11–14]. Geochemical mapping approaches have not only focused on mineral exploration but also on the concerns of the environment, human health, land use, and others. Thus, many elements are being given multipurpose applications. The China Geochemical Baselines (CGB) Project, with the determination of 76 elements carried out from 2008 to 2014 [15] is the only project to meet the requirements for the analysis of 71 chemical elements by the global geochemical baselines [16].

The National-Scale Geochemical Mapping Project in Lao People's Democratic Republic (PDR) (collectively, NGMPL) was supported financially and technically by China according to the agreement between China and Laos, implemented during 2015–2018. A total of 2079 stream sediment samples were collected at an average density of 1 sample/100 km² from the entire territory of Laos. In this paper, based on the statistics obtained by NGMPL, the Laos Geochemical Baseline (LGB) values of 69 elements are presented. We further discuss the effects of geological background, mineralization, hypergene processes, and human activities on the concentrations of these 69 elements through a comparison of the average crustal (CA) values [17,18] and CGB [19]. The baseline data are important for the evaluation and development of mineral resources, agriculture, and environment monitoring in Laos. The geochemical background values of 69 elements were compared with the crustal abundance and CGB values. The results show that under the influence of geological background, tectonics, magmatic activity and supergenesis, the results of statistical analysis of elemental parameters and R-type factor analysis have their own characteristics. The enrichment of elements has a good indicative significance for the deposits. The baseline data are of importance for the evaluation and development of mineral resources, agriculture, and environment monitoring in Laos.

2. Geological Background

Laos is located in the north-central part of the south-central Asian peninsula, with 80% of its territory occupied by mountains and the rest by basins and plains. It has a tropical and subtropical monsoon climate, with extremely developed vegetation, strong modern weathering and accumulation, and little bedrock exposure, which limits geological investigations due to inconvenient transportation. The strata exposed in Laos are mainly Paleozoic, followed by the Mesozoic and Cenozoic. The Paleozoic strata are mainly developed in the northern and eastern areas. The Mesozoic strata are mainly composed of Middle and Upper Triassic marine tuffs, sandstones, and siltstones exposed in the Xam Nua area, followed by Jurassic marine deposits in the southern valley. The Eocene strata developed in the northern intermountain valleys, and lignite seams are present; the Quaternary strata are widely distributed in the gullies. Tectonically, Laos belongs to the Indochina plate, which is a complex and unified land mass composed of many small blocks and interconnected zones. It is divided into seven tertiary geotectonic units from west to east (Figure 1): the Jinghong-Sukhothai, Simao-Phitsanulok, Vientiane-Kontum, Truong Son, Dien Bien Phu-Loei, Sepon-Tam Ky, and Ailaoshan-Song Ma [20–29].

The Jinghong-Sukhothai volcanic arc mainly develops Upper Carboniferous-Triassic terrigenous clastic rocks, tuffs and volcanic rocks. The geochemical data of Middle and Lower Triassic volcanic rocks show land margin arc features, and these early strata suffered late folding to form a NE folding zone and are covered by Late Triassic terrigenous sandstone unconformity. The intrusive rocks are mainly Early to Middle Triassic Island Arc and Late Triassic S granites [27]. The Simao-Phitsanulok block mainly consists of the Upper Triassic foreland basin-phase coal-bearing milarite sedimentary and Jurassic-Paleocene red terrestrial clastic rock system. The Upper Triassic-Cretaceous terrestrial salt-bearing red clastic rock system is unconformably overlain by the Carboniferous-Permian terrestrial clastic rock-carbonate rock and volcanic rock system [27,30,31]. The Vientiane-Kontum block is strongly magmatic, with granitic intrusive rock types mainly developed from Precambrian to Mesozoic. Large, rifted overflow basalts developed in Cenozoic, and the Mesozoic basin is mainly a set of Triassic-Paleocene terrestrial salt-bearing red-bedded rock systems. The Mekong fold belt is mainly composed of Paleozoic strata, developing a Cambrian-Silurian shallow metamorphic volcanic-sedimentary rock system. The Kontum block is an ancient land mass, composed of a crystalline basement and cover [27,30,31]. During the formation of the Truong Son block, magmatic activity was exceptionally strong, mainly developing Precambrian intermediate and advanced metamorphic rocks and Ordovician-Cretaceous sedimentary and igneous rocks. The mixed lithified hornblende interspersed with black cloud plagioclase and crystalline gneisses developed in the Yuan Paleogene; the

calc-alkaline volcanic-intrusive rocks of Devonian-Carboniferous active land margin-island arc type and granites of Early-Middle Permian collisional orogeny and Late Carboniferous-Permian are mainly developed in the Phanerozoic. Carbonatite-land-derived clastic sedimentary rock systems and island-arc calc-alkaline volcanic rock systems such as basalt and andesite [27,32]. The DienBienPhu-Loei suture is composed of metamorphic peridotites, mafic rocks, basal lava and radiolarian siliciclastic rocks from bottom to top. In Laos, ultramafic rocks are only found in the intersection of the DienBienPhu Fracture Zone and the Nam Kham Valley in Luang Prabang East [27,30,31]. The Sepon-TamKy suture is distributed in a north-western direction, and the serpentine green mixed rock assemblage represented by ultramafic, magnesian and terrestrial clastic rocks is exposed. The serpentine green mixed rock bodies are exposed intermittently along the suture zone in the form of tectonic lenses owing to late strong tectonic deformation [27,32]. A relatively complete ophiolite sequence is developed in the Ailaoshan-Song Ma procedure, with metamorphic peridotite, stacked crystal miscellaneous rocks, basal lava, and radiolarian-bearing siliciclastic rocks from the bottom to the top in order [27,30,33]. The magma belts are mostly located in subduction zones, collision zones, and other areas of strong plate tectonic activity, forming tectonic magma belts with geotectonic significance. These tectonic magma belts are closely related to the formation of mineral resources in the region.

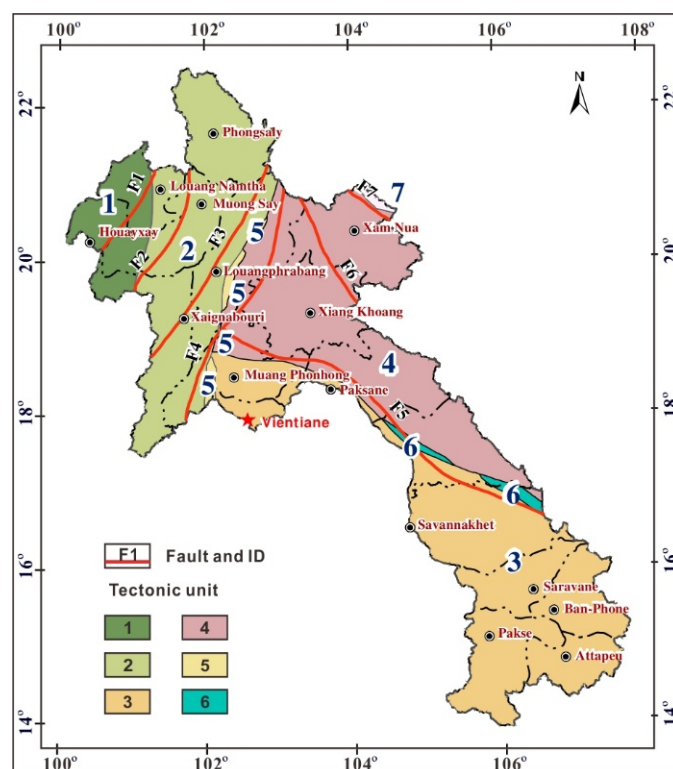


Figure 1. Tectonic sketch of Laos (modified from [27,28]). Tectonic units: 1—Jinghong-Sukhothai arc; 2—Simao-Phitsanulok block; 3—Vientiane-Kontum block; 4—Truong Son block; 5—DienBienPhu-Loei suture; 6—Sepon-TamKy suture; 7—Ailaoshan-Song Ma procedure. Faults: F1—Lancangjiang-Ban Namkham fault; F2—Nam Beng fault; F3—Louangphrabang fault; F4—Phu Pulei fault; F5—Truong Son-Da Nang fault; F6—Song Lan fault; F7—Song Ma fault.

3. Materials and Methods

3.1. Sampling

Sampling for the NGMPL took place during the period of 2015–2018. The sampling densities of geochemical mapping were dependent on the map scale. The guideline used for determining the sampling density in a geochemical mapping project was based on the map scale, along with the following criteria: each square-centimeter area should have at least

one datum on the map. For example, in a geochemical map generated at a 1:1,000,000 scale, each 1 cm × 1 cm area in the map is equal to one 10 km × 10 km field sampling cell. For GPS convenience and sample management, one sample will be taken from each 1:25,000 map sheet, i.e., each sampling cell is 7.5' (long.) by 5.0' (lat.), approximately 100 km² (Figure 2a). The guidelines used for designing the sampling layout were as follows: (1) sampling sites should be distributed as evenly as possible throughout the survey area; (2) each sample site should be at only one position, thereby allowing control of the maximum area of a sampling cell (Figure 2b).

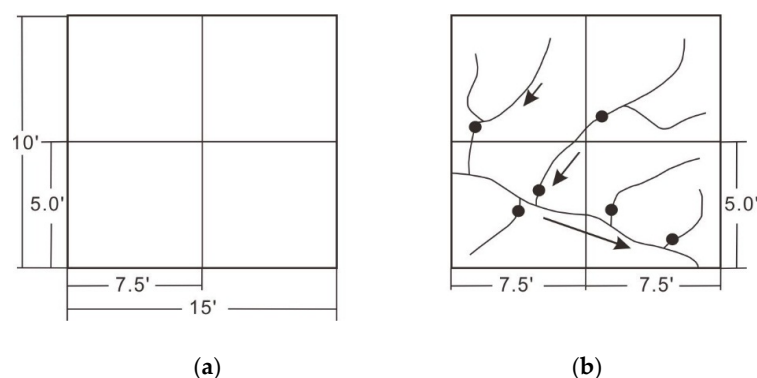


Figure 2. Grid cell (a) and sample location layout (b). Black arrows indicate the directions of the river.

The preference for sampling media was stream sediment, thus active stream sediment samples were taken at the riverbed surface at a depth range of 0–25 cm (excluding materials from the organic layer where present) in hilly and mountainous terrains. Regoliths or soils were the preferred sampling media from the top layer of soil horizon B, usually at a depth of 20–40 cm in plain terrains. The grain size of the samples was determined to be −10 mesh (2 mm), which can delineate the geochemical anomalies of key ore-prospecting elements [17]. Samples were sieved through −10 mesh on-site or in field camps after natural drying. In total, 2079 samples (including ~5% field duplicates) were collected during the NGML project (Figure 3).

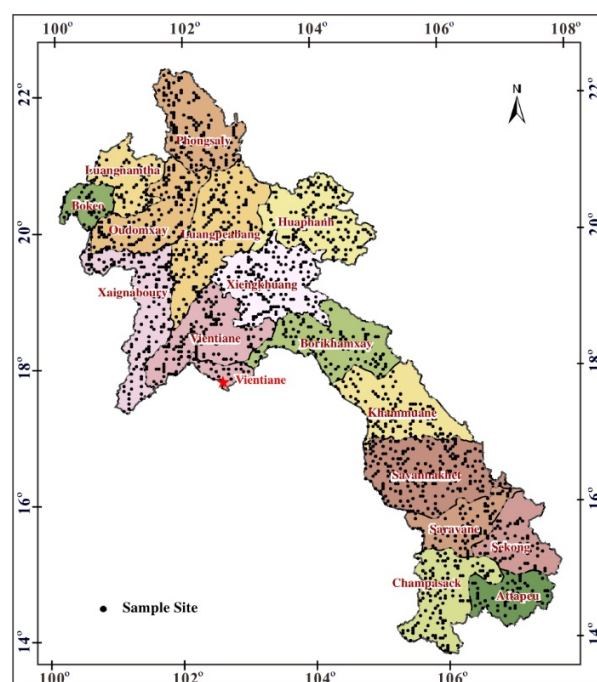


Figure 3. Sample sites of the National-scale Geochemical Mapping Project of Lao PDR (NGMPL).

3.2. Laboratory Chemical Analysis

The raw samples were dried in an oven at 45 °C in the laboratory. An 80 g sample was weighed and ground in an agate ball mill to a –200 mesh. The samples were analyzed for 69 elements using ICP-OES, ICP-MS, and XRF in combination with other methods: ICP-OES for seven elements (CaO, MgO, Na₂O, Be, Li, Mn, and Sr); ICP-MS for 29 elements (Bi, Cd, Cs, Hf, In, Mo, Pb, Sc, Ta, Te, Th, Tl, U, W, La, Y, Ce, Dy, Er, Eu, Gd, Ho, Lu, Nd, Pr, Sm, Tb, Tm, and Yb); XRF for 20 elements (SiO₂, Al₂O₃, Tot. Fe₂O₃, KO₂, Ba, Br, Cl, Co, Cr, Cu, Ga, Nb, Ni, P, Rb, S, Ti, V, Zn, and Zr); AFS for five elements (As, Hg, Sb, Se, and Ge); ES for Ag, B, and Sn; ISE for F; and GF-AAS for trace gold (Tables 1 and 2) [34]. Analytical accuracy and precision for laboratory quality were strictly controlled by laboratory replicate samples and standard reference materials [15].

Table 1. Analysis methods used for the NGMPL.

Analysis Method	Items	Elements Measured
Inductively coupled plasma optical emission spectrometry (ICP-OES)	7	CaO, MgO, Na ₂ O, Be, Li, Mn, Sr
X-ray fluorescence spectrometry (XRF)	20	SiO ₂ , Al ₂ O ₃ , Tot. Fe ₂ O ₃ , KO ₂ , Ba, Br, Cl, Co, Cr, Cu, Ga, Nb, Ni, P, Rb, S, Ti, V, Zn, Zr
Inductively coupled plasma mass spectrometry (ICP-MS)	29	Bi, Cd, Cs, Hf, In, Mo, Pb, Sc, Ta, Te, Th, Tl, U, W, La, Y, Ce, Dy, Er, Eu, Gd, Ho, Lu, Nd, Pr, Sm, Tb, Tm, Yb
Atomic fluorescence spectrometry (AFS)	5	As, Hg, Sb, Se, Ge
Emission spectrometry (ES)	3	Ag, B, Sn
Ion selective electrode (ISE)	1	F
Colorimetry (COL)	1	I
Oxidative combustion and gas chromatography (GC)	2	C, N
Graphite furnace/Flame atomic absorption spectrometry (GF-AAS)	1	Au

Table 2. Detection limits of the 69 elements. SiO₂, Al₂O₃, Tot. Fe₂O₃, MgO, CaO, Na₂O, K₂O, and C are expressed in %; Ag, Au, Cd, and Hg are expressed in µg/kg; other elements are expressed in mg/kg; LoD = limit of detection.

ID	Element	Minimum Requirement		ID	Element	Minimum Requirement	
		Analysis Method	LoD			Analysis Method	LoD
1	Ag	ES	20	36	Sr	ICP-OES	5
2	As	AFS	1	37	Ta	ICP-MS	0.1
3	Au	GF-AAS	0.2	38	Te	ICP-MS	0.01
4	B	ES	1	39	Th	ICP-MS	1.0
5	Ba	XRF	5	40	Ti	XRF	10
6	Be	ICP-OES	0.5	41	Tl	ICP-MS	0.1
7	Bi	ICP-MS	0.05	42	U	ICP-MS	0.1
8	Br	XRF	1.0	43	V	XRF	5
9	Cd	ICP-MS	20	44	W	ICP-MS	0.2
10	Cl	XRF	20	45	Zn	XRF	2
11	Co	XRF	1	46	Zr	XRF	2
12	Cr	XRF	5	47	Y	ICP-MS	1
13	Cs	ICP-MS	1	48	La	ICP-MS	1
14	Cu	XRF	1	49	Ce	ICP-MS	1
15	F	ISE	100	50	Pr	ICP-MS	0.1
16	Ga	XRF	2	51	Nd	ICP-MS	0.1
17	Ge	AFS	0.1	52	Sm	ICP-MS	0.1
18	Hf	ICP-MS	0.2	53	Eu	ICP-MS	0.1
19	Hg	AFS	2	54	Gd	ICP-MS	0.1
20	I	COL	0.5	55	Tb	ICP-MS	0.1
21	In	ICP-MS	0.02	56	Dy	ICP-MS	0.1

Table 2. Cont.

ID	Element	Minimum Requirement		ID	Element	Minimum Requirement	
		Analysis Method	LoD			Analysis Method	LoD
22	Li	ICP-OES	1	57	Ho	ICP-MS	0.1
23	Mn	ICP-OES	10	58	Er	ICP-MS	0.1
24	Mo	ICP-MS	0.2	59	Tm	ICP-MS	0.1
25	N	GC	20	60	Yb	ICP-MS	0.1
26	Nb	XRF	2	61	Lu	ICP-MS	0.1
27	Ni	XRF	2	62	SiO ₂	XRF	0.1
28	P	XRF	10	63	Al ₂ O ₃	XRF	0.05
29	Pb	ICP-MS	2	64	Tot. Fe ₂ O ₃	XRF	0.05
30	Rb	XRF	5	65	MgO	ICP-OES	0.05
31	S	XRF	30	66	CaO	ICP-OES	0.05
32	Sb	AFS	0.05	67	Na ₂ O	ICP-OES	0.05
33	Sc	ICP-MS	1	68	K ₂ O	XRF	0.05
34	Se	AFS	0.01	69	TC	GC	0.1
35	Sn	ES	1.0				

3.3. R-Mode Factor Analysis

Factor analysis is a statistical technique of data reduction used to explain the correlations between observed variables in terms of a smaller number of unobserved variables called factors [35–37]. In this study, an R-mode factor analysis, based on principal component analysis and varimax rotation, was performed on the database ($n = 2079$) to evaluate relationships among all the elements, and factors with eigenvalues >1 were extracted. Prior to the factor analysis, the BDL values were replaced with 0.5 of the detection limits. The centered log-ratio (clr) transformation proposed by Aitchison [38] was performed on the raw data using the CoDaPack [39,40], which is widely used for processing compositional data [41–43]. In addition, the clr-transformed data, rather than the raw data, was normally distributed and it satisfied the preconditions for factor analysis (Figure 4a–d).

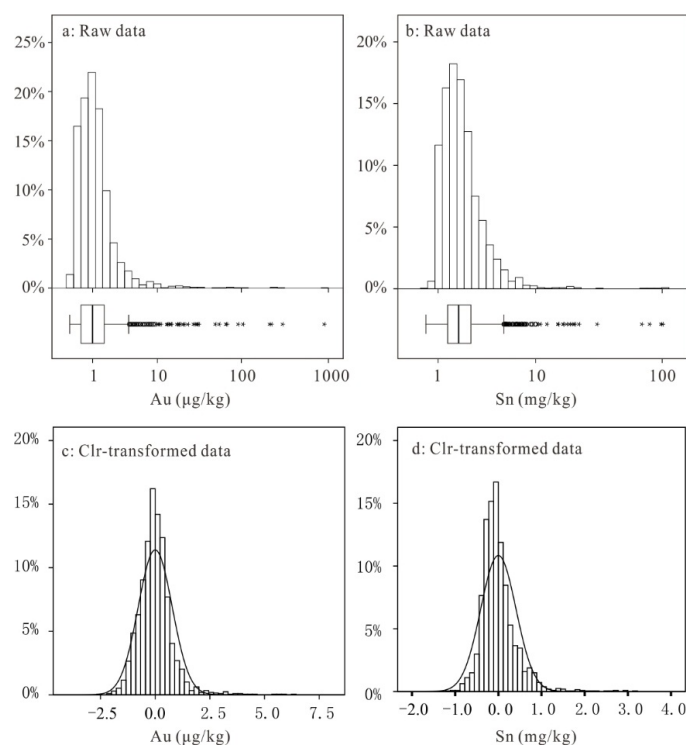


Figure 4. (a) Histogram and boxplot of Au on the raw data, (b) Histogram and boxplot of Sn on the raw data, (c) Histogram of Au on the clr-transformed data, (d) Histogram of Sn on the clr-transformed data. Asterisks indicate cases with values more than 1.5 times box length from the upper edge of the box.

4. Results

The concept of geochemical background, which originally emerged in exploration geochemistry [44–46], generally represents the average value in the natural levels (i.e., not affected by human activities). There are numerous ways to express background concentrations, e.g., arithmetic mean (AM), GM, median, upper confidence limit (UCL) of the mean, upper baseline concentration (UBC), and nth percentile [47]. However, the related soils or sediments are no longer “natural”, given that human-induced changes have historically occurred owing to agriculture and industry. A baseline is used when measuring present levels so that future changes can be recognized or quantified. The establishment of baseline values of chemical elements in soils or sediments is a critical issue [19,45]; as Zoback [48] states, “How do we recognize and understand changes in natural systems if we do not understand the range of baseline levels?” Laos is dominated by agriculture, but not much by modern industry. Therefore, the background is less affected by industry and human activities. The background is used in the context of measuring levels so that any future changes can be quantified. In this study, the median value was accepted as the LGB values of the 69 elements in the stream sediments collected in Laos because the median value is the most robust statistical value of the median trend [19,49].

Table 3 presents the statistical results. The LGB values of As, B, Br, Cs, Hf, Li, N, Pb, Sb, Zr, and SiO₂ are 1.3 times the average crustal values [17]. Ba, Be, Cl, Co, Cr, Cu, F, Ga, Mn, Mo, Ni, S, Sc, Sr, Ti, Tl, V, Zn, Eu, Al₂O₃, Tot. Fe₂O₃, MgO, CaO, and Na₂O are less than 0.7 times the average crustal values.

Table 3. Statistical parameters of the 69 elements and their comparisons with the CA and CGB from NGMPL. SiO₂, Al₂O₃, Tot. Fe₂O₃, MgO, CaO, Na₂O, K₂O, and C are expressed in %; Ag, Au, Cd, and Hg are expressed in µg/kg; other elements are expressed in mg/kg; Q: percentile of the data set (Q50: median); Min: minimum concentration; Max: maximum concentration; CA: crust average value [17,18]; CGB: China Geochemical Baselines value [19]; LGB/CA = Q50 (median)/CA; LGB/CGB = Q50 (median)/CGB.

Element	LGB Values of 69 Elements						CA	CGB	Enrichment/Depletion Coefficient	
	Min.	Q25	Q50	Q75	Q85	Max.			LGB/CA	LGB/CGB
Ag	19	50	61	75	87	10,000	56	77	1.1	0.8
As	1	2	5	9	12	2755	3	9	1.7	0.6
Au	0.1	0.6	1.0	1.5	2.0	914.0	1.3	1.3	0.8	0.8
B	2	21	34	52	63	334	11	43	3.1	0.8
Ba	11	151	243	345	421	4673	456	512	0.5	0.5
Be	0.1	0.8	1.2	1.7	2.0	7.1	1.9	2.0	0.6	0.6
Bi	0.03	0.10	0.18	0.29	0.36	46.62	0.18	0.30	1.0	0.6
Br	0.1	1.1	1.5	2.1	2.8	36.6	0.9	2.2	1.7	0.7
Cd	1	37	63	107	150	41,650	80	137	0.8	0.5
Cl	21	51	61	73	81	5615	244	78	0.3	0.8
Co	<1	5	9	14	18	243	27	11	0.3	0.8
Cr	2	26	43	62	78	24,101	135	53	0.3	0.8
Cs	<1	2	3	5	7	26	2	6	1.5	0.5
Cu	1	10	16	26	33	459	27	20	0.6	0.8
F	21	182	271	374	441	2717	553	488	0.5	0.6
Ga	1	7	10	14	16	44	16	15	0.6	0.7
Ge	0.3	1.1	1.3	1.4	1.5	2.5	1.3	1.3	1.0	1.0
Hf	0.8	3.9	6.1	9.8	12.4	76.2	3.7	6.5	1.6	0.9
Hg	3	18	29	50	68	806	30	26	1.0	1.1
I	0.2	0.5	0.7	1.2	1.8	32.6	0.7	1.1	1.0	0.6
In	<0.01	0.03	0.04	0.06	0.07	10.16	0.05	0.05	0.8	0.8
Li	1	14	22	30	35	134	16	30	1.4	0.7
Mn	75	262	391	622	809	14,212	774	569	0.5	0.7
Mo	0.1	0.3	0.4	0.7	0.9	25.4	0.8	0.7	0.5	0.6
N	67	266	411	725	989	5290	56	707	7.3	0.6
Nb	<1	6	9	12	14	64	8	13	1.1	0.7
Ni	1	9	15	24	30	237	59	24	0.3	0.6
P	42	199	312	439	526	4157	436	570	0.7	0.5
Pb	2	10	15	22	27	4695	11	22	1.4	0.7

Table 3. Cont.

Element	LGB Values of 69 Elements						CA	CGB	Enrichment/Depletion Coefficient	
	Min.	Q25	Q50	Q75	Q85	Max.			LGB/CA	LGB/ CGB
Rb	3	31	52	83	103	381	49	96	1.1	0.5
S	19	52	72	113	153	2346	404	245	0.2	0.3
Sb	0.05	0.30	0.50	0.88	1.23	1284.94	0.20	0.73	2.5	0.7
Sc	<1	5	8	12	14	49	22	10	0.4	0.8
Se	0.02	0.06	0.10	0.19	0.25	3.17	0.13	0.17	0.8	0.6
Sn	0.6	1.4	1.9	2.5	3.2	100.0	1.7	3.0	1.1	0.6
Sr	1	25	43	66	82	584	320	197	0.1	0.2
Ta	0.1	0.5	0.7	1.0	1.1	5.4	0.7	1.0	1.0	0.7
Te	<0.01	0.02	0.04	0.04	0.05	1.25	–	0.04	–	1.0
Th	0.3	3.6	6.0	9.3	11.7	51.1	5.6	11.0	1.1	0.5
Ti	229	1921	2888	4009	4771	32,015	4200	3498	0.7	0.8
Tl	<0.1	0.2	0.3	0.5	0.7	2.6	0.5	0.6	0.6	0.5
U	0.2	1.0	1.5	2.3	2.8	17.8	1.3	2.5	1.2	0.6
V	4	36	59	89	109	569	138	70	0.4	0.8
W	0.1	0.6	0.9	1.4	1.8	37.2	1.0	1.6	0.9	0.6
Zn	4	28	47	66	77	2524	72	66	0.7	0.7
Zr	23	119	188	268	323	1886	132	230	1.4	0.8
Y	2	11	16	23	26	340	19	24	0.8	0.7
La	2	14	20	28	33	319	20	33	1.0	0.6
Ce	4	25	36	53	63	385	43	64	0.8	0.6
Pr	0.5	3.2	4.5	6.6	7.7	57.7	4.9	7.6	0.9	0.6
Nd	1.7	11.9	17	24.1	28.3	177.6	20.0	28.2	0.9	0.6
Sm	0.3	2.3	3.3	4.7	5.5	37.6	3.9	5.3	0.8	0.6
Eu	0.1	0.5	0.7	1.0	1.2	7.8	1.1	1.1	0.6	0.6
Gd	0.3	2.1	3.1	4.3	4.9	34.9	3.7	4.6	0.8	0.7
Tb	0.1	0.4	0.5	0.7	0.8	6.1	0.6	0.8	0.8	0.6
Dy	0.3	2.0	3.0	4.2	4.8	38.3	3.6	4.5	0.8	0.7
Ho	0.1	0.4	0.6	0.8	0.9	7.4	0.8	0.9	0.8	0.7
Er	0.2	1.2	1.7	2.4	2.7	20.3	2.1	2.5	0.8	0.7
Tm	<0.1	0.2	0.3	0.4	0.4	2.8	0.3	0.4	1.0	0.8
Yb	0.2	1.2	1.7	2.4	2.7	16.7	1.9	2.6	0.9	0.7
Lu	<0.1	0.2	0.3	0.4	0.4	2.4	0.3	0.4	1.0	0.8
ΣLREE	9.16	56.68	81.09	117.89	138.88	984.62	92.90	139.20	0.9	0.6
ΣHREE	2.79	18.42	27.19	38.13	43.71	469.14	32.25	40.70	0.8	0.7
ΣREE	11.95	75.10	108.28	156.02	182.95	1453.76	125.15	179.90	0.9	0.6
SiO ₂	12.1	69.3	76.4	82.8	85.7	94.3	60.7	66.7	1.3	1.1
Al ₂ O ₃	0.55	6.50	10.21	12.89	14.42	27.72	15.88	11.90	0.6	0.9
Tot. Fe ₂ O ₃	0.20	2.10	3.60	5.41	6.56	29.09	7.59	4.20	0.5	0.9
MgO	0.02	0.39	0.60	0.89	1.09	7.00	4.72	1.43	0.1	0.4
CaO	0.01	0.15	0.24	0.46	0.78	42.83	6.39	2.74	0.0	0.1
Na ₂ O	0.03	0.20	0.41	0.65	0.82	3.31	3.10	1.75	0.1	0.2
K ₂ O	0.07	0.80	1.30	1.95	2.36	6.88	1.79	2.36	0.7	0.6
TC	<0.1	0.2	0.4	0.8	1.2	10.0	–	1.3	–	0.3

Further, the LGB values of Ag, Au, Bi, Cd, Ge, Hg, I, In, Nb, P, Rb, Se, Sn, Ta, Th, U, W, Y, La, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and K₂O are 0.7–1.3 times of the average crustal values (Table 4 and Figure 5).

The LGB values of As, B, Br, Cs, Hf, Hg, I, N, Sb, Se, Sn, and W are 1.3 times of the CGB values [17], while that of Ba, Cl, Co, Cr, Cu, F, Ga, Mn, Ni, P, S, Sc, Sr, Te, V, Zn, La, Ce, Pr, Nd, Sm, Eu, Al₂O₃, Tot. Fe₂O₃, MgO, CaO, Na₂O, and K₂O are 0.3 times of the CGB values. The CGB values of Ag, Au, Be, Bi, Cd, Ge, In, Li, Mo, Nb, Pb, Rb, Ta, Th, Ti, Tl, U, Zr, Y, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, SiO₂, and C are 0.7–1.3 times of the CGB values [19] (Table 4 and Figure 5). Kaiser–Meyer–Olkin (KMO) and Bartlett’s sphericity tests were adopted to evaluate the adequacy of the data set for factor analysis. A KMO value of 0.927 (greater than the recommended minimum value of 0.5) and the significance of Bartlett’s sphericity test ($\chi^2 = 271,391.511$, $p = 0.000$) indicate the effectiveness of factor analysis and correlations among the variables. The factor analysis showed that the 69 elements fell into 12 factors explaining 77.93 % of the total variability (Table 5). Factor 1 explaining the largest proportion (11.02 %) of total variability, consists of positive loadings (Bi, Th, U, W, Cs, K₂O, Rb, and Tl), which indicate potassium salt deposits and negative loadings (V, Cr, Cu, Ni, Co, Ti, and

Tot. Fe₂O₃), which indicate the weathering of basic volcanic rocks. Factor 2 (accounting for 8.83 % of the variance) consists of positive loadings (Er, Tm, Ho, Yb, Lu, Y, Dy, Tb, and Gd), which indicate bedrocks rich in heavy rare earth. Factor 3 (accounting for 6.55 % of the variance) consists of positive loadings (SiO₂, Ge, Cl, Ag, and Zr), which indicate Ag-related mineralization. Factor 4 (accounting for 5.54 % of the variance) consists of positive loadings (La, Ce, Pr, Nd, Sm, and Gd), which indicate bedrock rich in light rare earth. Factor 5 (accounting for 5.07 % of the variance) consists of positive loadings (TC, N, S, Br, I, and Se), which may be related to farming activities. Factor 6 (accounting for 3.15% of the total variance) consists of positive loadings (Sr and Na₂O), which may be related to biogenic products and depositional conditions besides source rock characteristics [50,51]. Factor 7 (Ta-Nb association) and Factor 8 (Li-B association), accounting for 3.14% and 2.91% of the total variance, respectively, indicate their highly similar geochemical behaviors. Factor 9 (Sb-As association), accounting for 2.34% of the total variance, indicates hydrothermal activities and possibly serves as an indicator of mineral exploration. Factor 10 (Al₂O₃), accounting for 2.24% of the total variance, indicates Al₂O₃-rich areas caused by strong weathering. Factor 11 (Mo) and Factor 12 (Au), accounting for 1.57% and 1.14% of the total variance, respectively, indicate Mo- and Au-related mineralization, respectively.

Table 4. Statistical parameter enrichment/depletion coefficient of the 69 elements with the CA and CGB from NGMPL. SiO₂, Al₂O₃, Tot. Fe₂O₃, MgO, CaO, Na₂O, K₂O, and C are expressed in %; Ag, Au, Cd, and Hg are expressed in µg/kg; other elements are expressed in mg/kg; Q: percentile of the data set (Q50: median); Min: minimum concentration; Max: maximum concentration; CA: crust average value [17,18]; CGB: China Geochemical Baselines value [19]; LGB/CA = Q50 (median)/CA; LGB/CGB = Q50 (median)/CGB.

	Depletion	0.7–1.3	Enrichment
	<0.7		>1.3
LGB/CA	Ba, Be, Cl, Co, Cr, Cu, F, Ga, Mn, Mo, Ni, S, Sc, Sr, Ti, Tl, V, Zn, Eu, Al ₂ O ₃ , Tot. Fe ₂ O ₃ , MgO, CaO, Na ₂ O	Ag, Au, Bi, Cd, Ge, Hg, I, In, Nb, P, Rb, Se, Sn, Ta, Th, U, W, Y, La, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, K ₂ O	As, B, Br, Cs, Hf, Li, N, Pb, Sb, Zr, SiO ₂
LGB/CGB	Ba, Cl, Co, Cr, Cu, F, Ga, Mn, Ni, P, S, Sc, Sr, Te, V, Zn, La, Ce, Pr, Nd, Sm, Eu, Al ₂ O ₃ , TFe ₂ O ₃ , MgO, CaO, Na ₂ O, K ₂ O	Ag, Au, Be, Bi, Cd, Ge, In, Li, Mo, Nb, Pb, Rb, Ta, Th, Ti, Tl, U, Zr, Y, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, SiO ₂	As, B, Br, Cs, Hf, Hg, I, N, Sb, Se, Sn, W

Table 5. Results of the factor analysis based on the clr-transformed data and the selected factors based on threshold of 0.6 (the absolute threshold value).

Factor	Element Association	Relations	Eigenvalue	% of Variance	Cumulative %
Factor 1	Bi-Th-U-W-Cs-K ₂ O-Rb-Tl-(V-Cr-Cu-Ni-Co-Ti-Tot. Fe ₂ O ₃)	Acid rocks, Basic volcanic rocks	11.02	15.98	15.98
Factor 2	Er-Tm-Ho-Yb-Lu-Y-Dy-Tb-Gd	Bedrocks	8.83	12.79	28.77
Factor 3	SiO ₂ -Ge-Cl-Ag-Zr	Mineralizations	6.55	9.49	38.26
Factor 4	La-Ce-Pr-Nd-Sm-Gd	Bedrocks	5.54	8.03	46.29
Factor 5	TC-N-S-Br-I-Se	Farming activities	5.07	7.35	53.64
Factor 6	Sr-Na ₂ O	Bedrocks	3.15	4.57	58.20
Factor 7	Ta-Nb	Bedrocks	3.14	4.55	62.75
Factor 8	Li-B	Bedrocks	2.91	4.22	66.97
Factor 9	Sb-As	Mineralizations	2.34	3.39	70.36
Factor 10	Al ₂ O ₃	Bedrocks	2.24	3.25	73.61
Factor 11	Mo	Mo mineralizations	1.57	2.27	75.88
Factor 12	Au	Au mineralizations	1.41	2.05	77.93

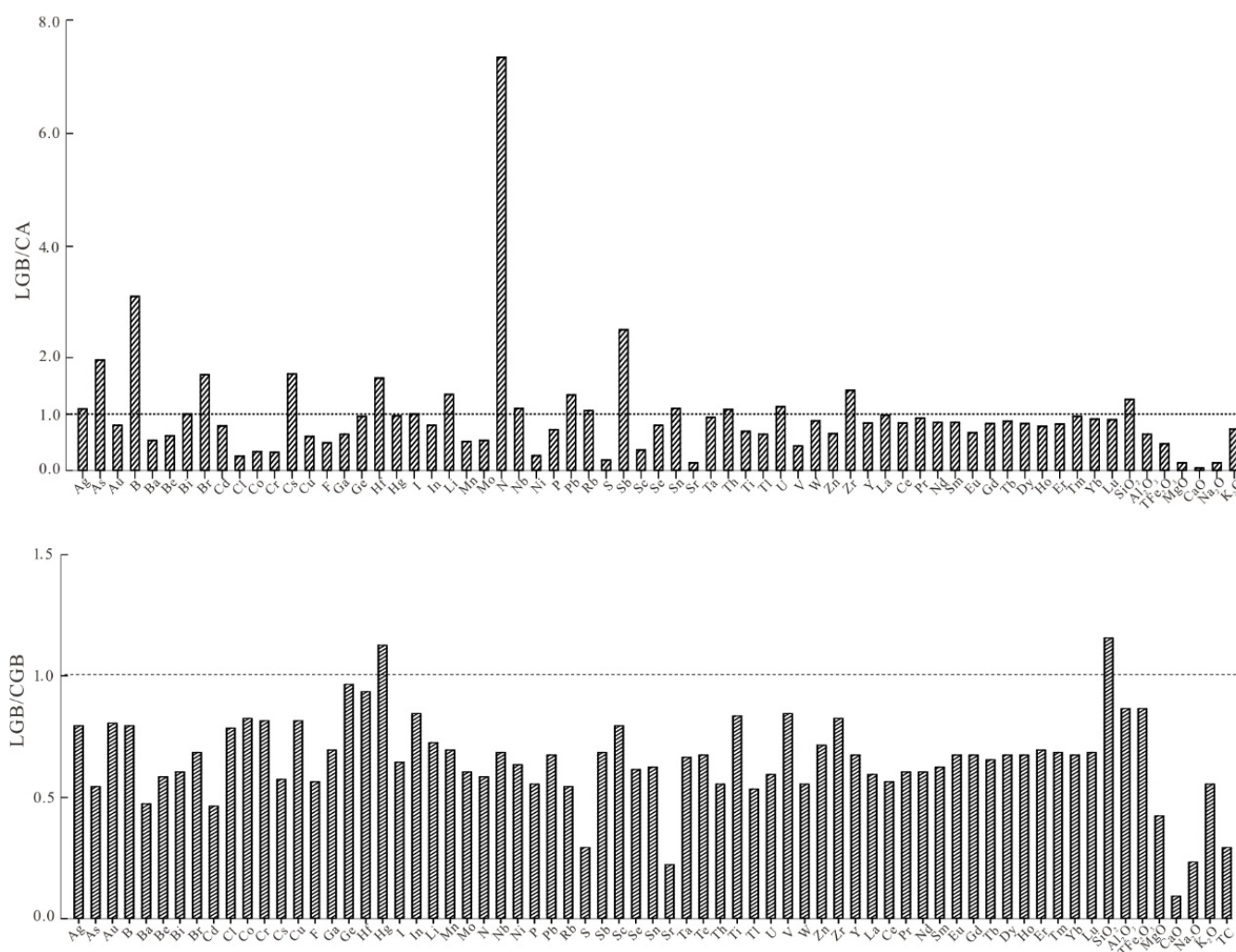


Figure 5. Bar graph showing the enrichment/depletion of the 69 elements using the ratio of the median and the CA (upper) and CGB (top) values.

5. Discussion

5.1. Elements Related to Bedrock

The enrichment/depletion coefficients of the major elements are ranked as follows: $\text{SiO}_2 > \text{K}_2\text{O} > \text{Al}_2\text{O}_3 > \text{TFe}_2\text{O}_3 > \text{MgO} > \text{Na}_2\text{O} > \text{CaO}$ (Table 3 and Figure 5). The concentrations of major elements are controlled by both bedrock and weathering processes. All the major elements of SiO_2 in stream sediments were enriched, whereas CaO was significantly depleted. SiO_2 content in stream sediment shows a pronounced variability from 12.07 to 94.34%, with a median of 76.43%, which is related to the bedrock geology (sandstone, granite, granodiorite and their metamorphosed equivalents), and also to quartz-rich superficial deposits. Zhu [23] reported a large area of Mesozoic sandstone in northwestern and southern Laos, and a major granite area in northern Laos. The enrichment of SiO_2 is related to the residue of quartz-rich sand (while the other elements are leached and bedrocks of sandstone and granites). During the supergenesis of heavy precipitation with an average annual precipitation of 3700 mm in the subtropical region, Ca, Mg, and Na leached, and the median values were significantly lower than that of the crustal abundance. The significantly depleted CaO value when compared with the average crustal and CGB value is contributed by the dissolution of carbonate minerals. The median value of Al_2O_3 is 10.21% with a range of 0.55% to 27.72%. The enrichment of Al_2O_3 is mainly caused by the weathering of carbonate and basaltic rocks. Bauxite deposits were reported by Zhu [23] mainly distributed in the Boloven Plateau, which is located in southern Laos. The mining

area is of high temperature and rainy, the physical weathering of the source layer, especially the chemical weathering and biological weathering, is very strong, the plateau topography is gentle, there is no fault fold in the area, the structure is simple, suitable for the formation of bauxite. It is known that bauxite belongs to Lateritic bauxite. It is a high-quality lateritic gibbsite bauxite deposit formed in a low-lying area after basalt weathering.

There is a negative correlation between the Rb-K₂O-Tl-Cs-Th-Be-Ba-U-W association and the V-Cr-Cu-Ni-Co-Ti-TFe₂O₃ association in Factor 1 (Figure 6). The V-Cr-Cu-Ni-Co-Ti-TFe₂O₃ association represents the weathering of basic volcanic rocks, which were originally depleted in Bi, Th, U, W, Cs, K₂O, Rb, and Tl; weathering further leaches these elements from the basic volcanic rocks. However, the potassium salt deposits are strongly depleted in V, Cr, Cu, Ni, Co, Ti, and TFe₂O₃ and enriched in Bi, Th, U, W, Cs, K₂O, Rb, and Tl [52,53].

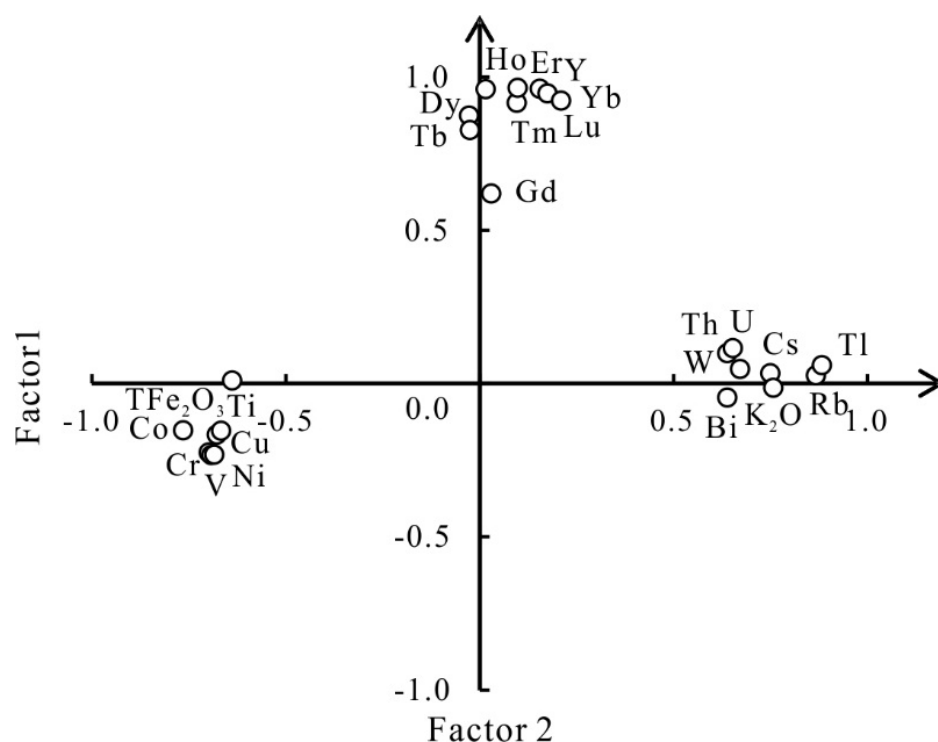


Figure 6. Biplot of Factor 1 (Bi–Th–U–W–Cs–K₂O–Rb–Tl–(V–Cr–Cu–Ni–Co–Ti–TFe₂O₃)) vs. Factor 2 (Er–Tm–Ho–Yb–Lu–Y–Dy–Tb–Gd association). Circles indicate elements.

5.2. Rare Earth Elements

The baseline values of rare earth elements (REEs) (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and Y) are generally consistent with the average crustal values (Table 3 and Figure 5). The REEs exhibit light rare earth element (LREE) enrichment and heavy rare earth element (HREE) depletion, with a negative Eu anomaly curve. The distribution pattern is similar to that of the CGBs with LREE enrichment, flat HREE distributions, and a negative Eu anomaly (Figure 7). The negative Eu anomaly may be due to the leaching of Eu with Ca, in which CaO is significantly depleted in the stream sediments mentioned above; the isomorphism of Eu and Ca exists when the Eu valence state is +2. The results of the factor analysis are consistent with the REE patterns. The LREEs (La, Ce, Pr, Nd, and Sm) and HREEs (Tb, Dy, Ho, Er, Tm, Yb, Lu, and Y) belong to Factors 4 and 2, respectively; Gd belongs to factors 2 and 4, because the loadings of Gd in both factors 2 and 4 are high, 0.62 and 0.71, respectively; and Eu does not belong to any factor (Figure 8).

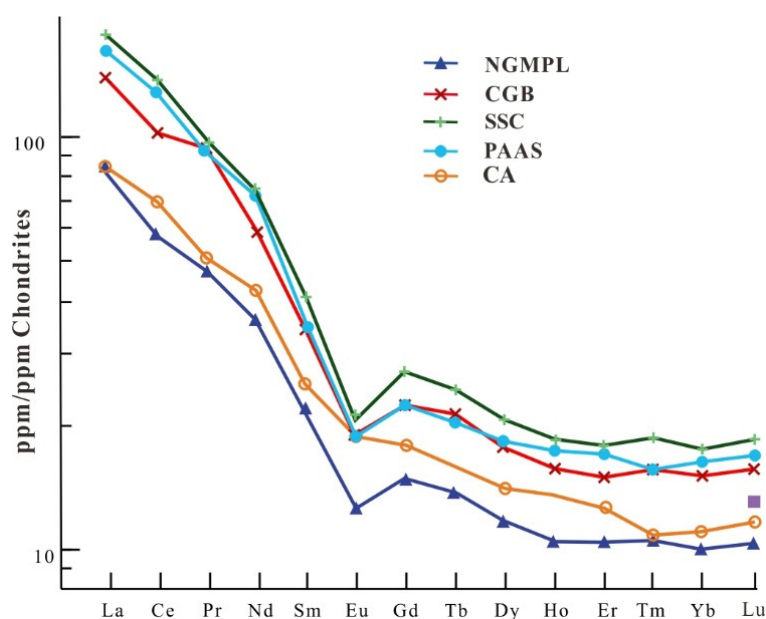


Figure 7. Chondrite-normalized REEs diagrams. (Chondrite and PAAS data after [54,55]). REEs, LREEs, HREEs are expressed in mg/kg; N: chondrites; NGMPL: The National-Scale Geochemical Mapping Project in Lao PDR; CGB: China Geochemical Baselines value [19]; SSC: Stream sediment in South China [56]; PAAS: Australian Post-Archean average shale [55]; CA: Average crustal values [17].

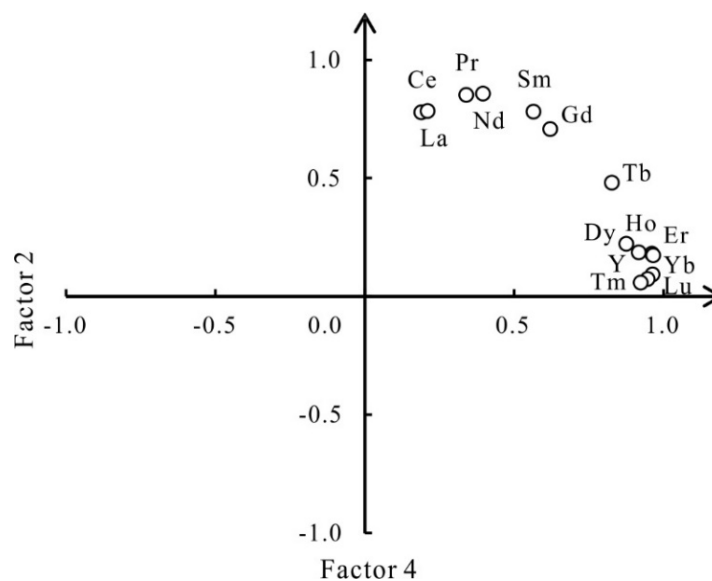


Figure 8. Biplot of Factor 2 vs. Factor 4 indicating the fractionation of LREEs and HREEs. Circles indicate elements.

The low concentrations of REEs in stream sediments are caused by quartz dilution as well as REEs activation during warm and wet weathering [55,57–59]. The ratio of LREEs to HREEs is consistent with that of the average crustal value and slightly lower than that of the CGB value (Table 6).

In tropical terrains, the ions absorbed into clays tend to feature more HREEs than LREEs [60]. Thus, HREEs account for a higher percentage of REEs in Laos' tropical zone than in China's temperate and subtropical zones. The areas with high concentrations of REEs are in the north on the border between China and Laos, which is consistent with new anomalies delineated and ion-adsorption type REE deposits discovered in Yunnan

Province, China [60,61]. This extension of the rare earth metallogenic belt may host ion-adsorption type REEs deposits that are distributed along the Truong Son-Da Nang fault, and the spatial distribution correlates with the granite distribution in the Truong Son block and its intersection area with the Sepon–TamKy suture zone. These findings hold promise for the subsequent exploration of rare earth minerals in Laos.

Table 6. Rare earth element characteristic parameter table.

	Σ REE (mg/kg)	Σ LREE (mg/kg)	Σ HREE (mg/kg)	L/H	δ Eu	δ Ce	(La/Sm) _N	(Gd/Yb) _N	(La/Yb) _N
NGMPL	108.28	81.09	27.19	2.98	0.71	0.94	3.80	1.47	8.09
CGB	179.90	139.20	40.70	3.42	0.68	0.99	4.02	1.46	9.10
Crust average	125.15	92.90	32.25	2.88	0.89	1.06	3.31	1.61	7.55

5.3. Elements Related to Mineralization

Au and Sn are the most important mineral commodities in Laos. The baselines of Au (1.04 µg/kg) are lower than the average crustal values (1.3 µg/kg) (Table 3 and Figure 5). It seems that the depletion of Au is not consistent with the rich gold deposits of Laos. The histogram graph (Figure 4a,b) shows that gold had abnormal distributions, and the boxplots indicate a certain number of samples distributed over Q75, which implies that the high concentrations of gold may be related to gold mineralization. The R-mode factor analysis indicated that Au was represented only in Factor 12, suggesting the absence of significant correlations between Au and other elements (Table 5 and Figure 9). Gold deposits are widely distributed in Laos and are mainly of alluvial, porphyry, quartz vein, and Carlin types. Residual and alluvial-flood gold deposits are the most numerous and widely distributed in Laos. Alluvial gold deposits are mainly concentrated in northern Louangphrabang, the southwest border including Thailand, and the area of Ban Thong kha in the central part of the country. Most of the rivers eroded the pre-Mesozoic metamorphic strata intruded by Mesozoic rocks, and the gold may be associated with the gold-bearing quartz veins in the metamorphic rocks [7,23,26,29,62]. Elemental compositions of different types of gold deposits are quite distinct, and even elemental composition of the same type of gold ore deposits may have a large difference; hydrothermal gold deposits may have high concentrations of volatile elements and metal sulfides, while alluvial-flood gold deposits may have high concentrations of platinum group elements and heavy minerals [63–70]. In addition, Au is highly active in supergene environments. Gold in primary Au deposits can experience mobilization, migration, and reprecipitation under the action of physical, chemical, and biogeochemical processes [71–75]. The diversity of gold ore types and the complexity of metallogenic processes in Laos, and the high mobility of gold in supergene environments indicate no obvious correlation between gold and other elements.

The high concentrations of Sn are highly consistent with the distribution of tin deposits in Laos. The Nam Pathene tin-high concentration area in Khammuane province is the largest tin mine-producing area in Laos [29] and the mine is located in the Nam Pathene area, tectonically at the intersection of the Truong son block and the Sepon–TamKy suture, extending the Truong son–Da Nang fault [23]. The types of tin ore deposits are predominantly alluvial, followed by skarn and hydrothermal.

The study shows that intense hydrothermal activities play an important role in the formation of minerals such as Cu, Pb, and Zn [76]. However, the factor analysis failed to characterize the significance of these elements; Pb, Zn, and Sn were absent from all the factors which may be caused by the complexity of the metallogenic process and bedrocks, and the significant differences in their geochemical behaviors in the supergene environment. (Table 5 and Figure 10). Wang et al.'s [77] study shows that Cu in Laos is unevenly distributed; generally, it is high in the north and low in the south. Most of the copper deposits in Laos are skarn-type, hydrothermal or porphyry types, which are mainly controlled by

magmatic hydrothermal and tectonic conditions [25,78]. Most of the Pb-Zn deposits belong to the hydrothermal type, which was formed chiefly during the Late Mesozoic period [76].

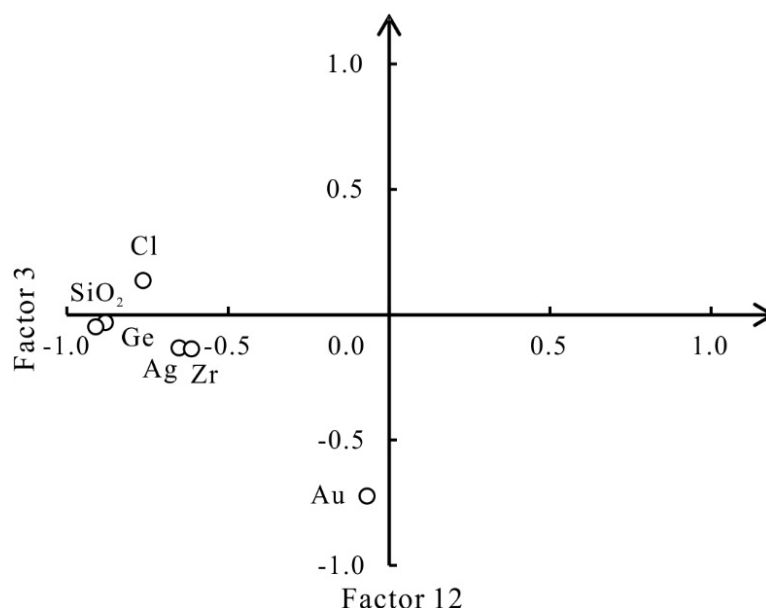


Figure 9. Biplot of Factor 3 (SiO_2 –Ge–Cl–Ag–Zr association) vs. Factor 12 (Au). Circles indicate elements.

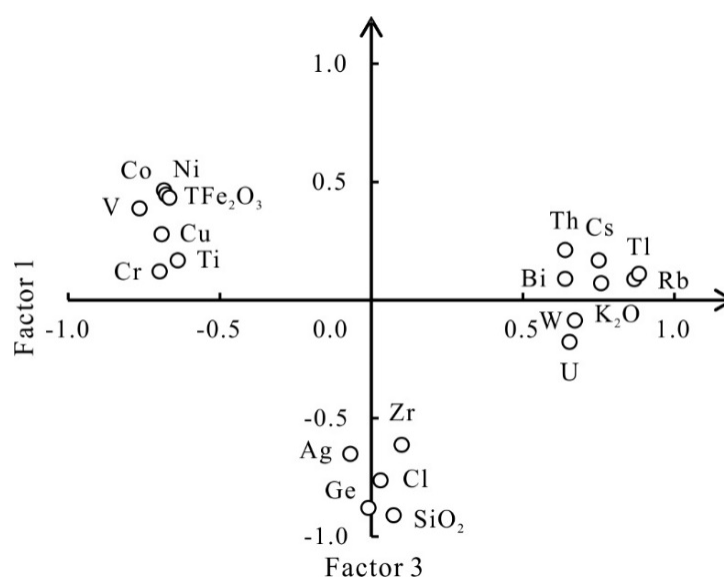


Figure 10. Biplot of Factor 1 (Bi–Th–U–W–Cs–K₂O–Rb–Tl–(V–Cr–Cu–Ni–Co–Ti–TFe₂O₃) association) vs. Factor 3 (SiO_2 –Ge–Cl–Ag–Zr association). Circles indicate elements.

The factor analysis results showed mineralization-related element assemblages (Factor 9: Sb–As association). The baseline values of Sb and As (0.5 and 4.9 mg/kg, respectively) were much higher than the average crustal values ($\text{RCC} > 1.3$); they were significantly enriched. Higher As concentrations (>11.7 mg/kg) are mostly influenced by lithology, mineralization, mining, and smelting activities. The study of [79,80] showed that As could be derived from pyrite. Zhu [23] reported a large area of contact with metasomatic and hydrothermal iron deposits in northern and central Laos. Pyrite and other metal sulfides are unevenly distributed in some ores. High concentrations of Sb (1.23 mg/kg) in stream sediment occur with Au–Cu–Sn–Sb–Pb mineralization. The richest Sb occurs in the Nam

Paten tin deposit area with 1285 mg/kg of Sb [29]. The enrichment of Sb and As in stream sediments can be an indicator of tin mineral deposits.

5.4. Elements Impacted by Human Activities

The elements C, N, P, and toxic metals, such as Hg and Cd, were found to be easily impacted by human activities. The median of N (411 mg/kg) was much higher than the average crustal value (56 mg/kg) with the most significant enrichment. Figure 11a represents the correlation between N and C. The scatter diagram indicates that most of the N and C samples fall in the same field of farming activities and a small proportion of samples in the field demonstrate a good correlation with CaO (Figure 11d), which is contributed by the limestone composition of calcite minerals (CaCO_3). Phosphorus has a good correlation with N and C, indicating that the concentration of N, C, and P are mainly impacted by human activities [53,81] (Figure 11b,c).

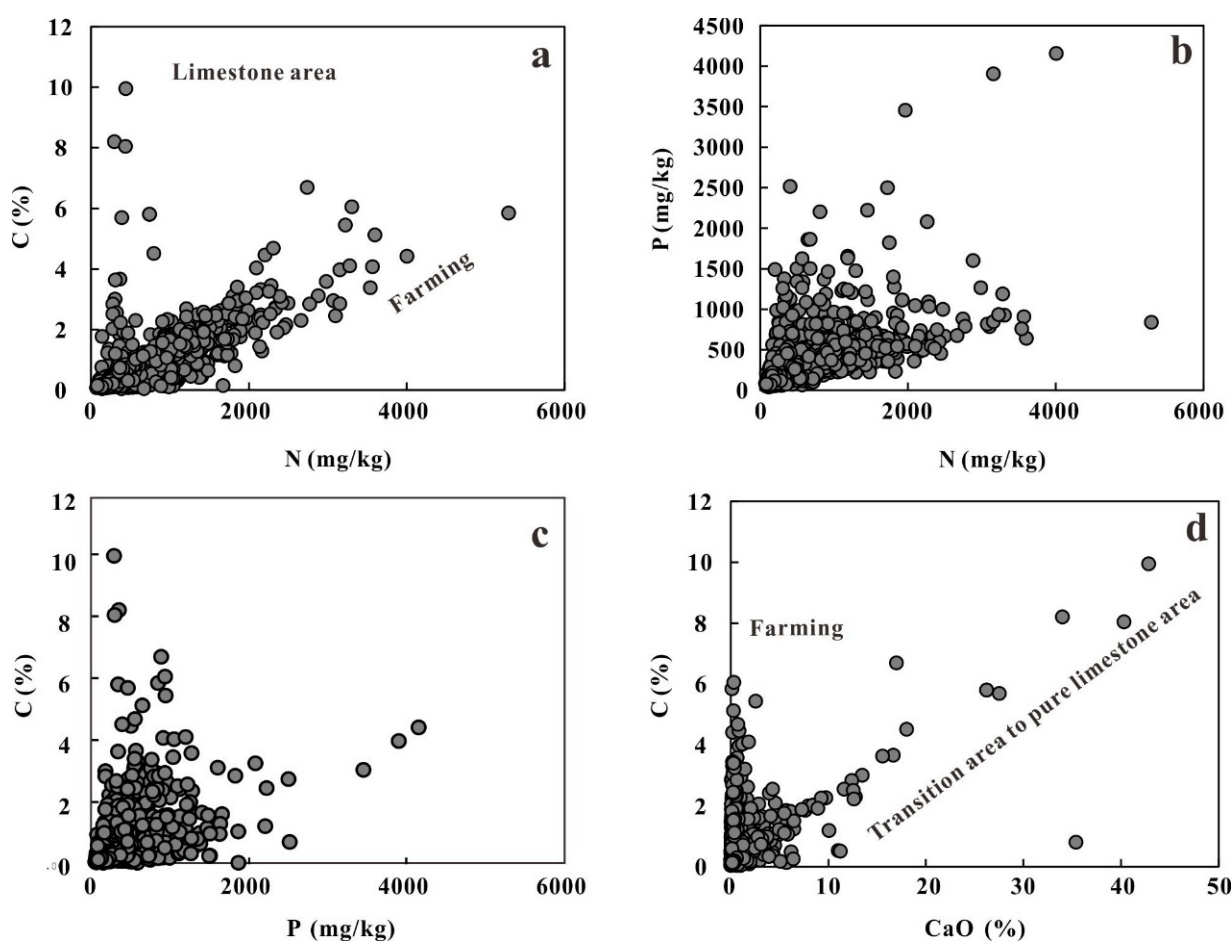


Figure 11. (a) Correlation scatter diagram between C and N, (b) Correlation scatter diagram between P and N, (c) Correlation scatter diagram between C and P, (d) Correlation scatter diagram between C and CaO.

The baseline value of Hg (29.1 $\mu\text{g/kg}$) is basically consistent with the average crustal value (30.0 $\mu\text{g/kg}$). High concentrations of Hg (>67.8 $\mu\text{g/kg}$) are mainly distributed in northern Laos as a result of gold mining activities that use Hg to extract Au (reference) [29]. There is a long history of gold mining and processing in Laos, which has caused Hg pollution in the Mekong River and its tributaries and has added a certain degree to the Hg concentration in the surrounding environment [20,82]. The enriched concentrations of mercury are also related to Au, Cu, Pb-Zn, and other minerals along the Louangphrabang, Nam Beng Truong, and Son-Da Nang faults [83].

5.5. Halogen Elements Linked to Climate

The coefficient values of the halogen elements are ranked as follows (Table 3 and Figure 5): $\text{Br} > \text{I} > \text{F} > \text{Cl}$. The baseline value of Br (1.5 mg/kg) is higher than the average crustal value (0.88 mg/kg). The diagram represents a positive correlation between Br and C (Figures 12 and 13).

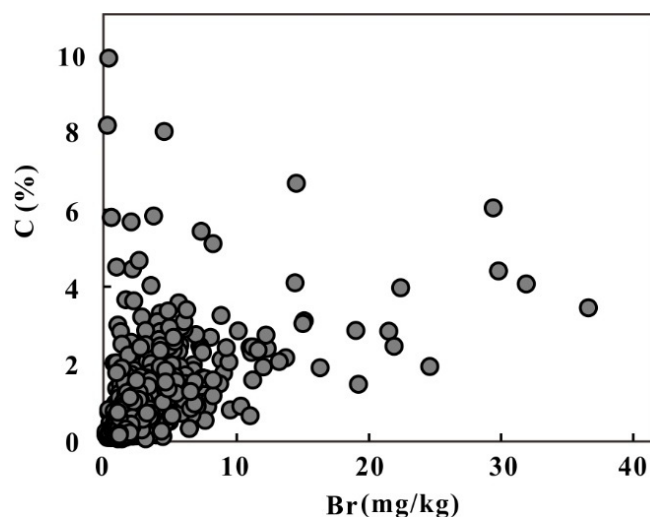


Figure 12. Correlation of C and Br contents in stream sediments from NGMPL.

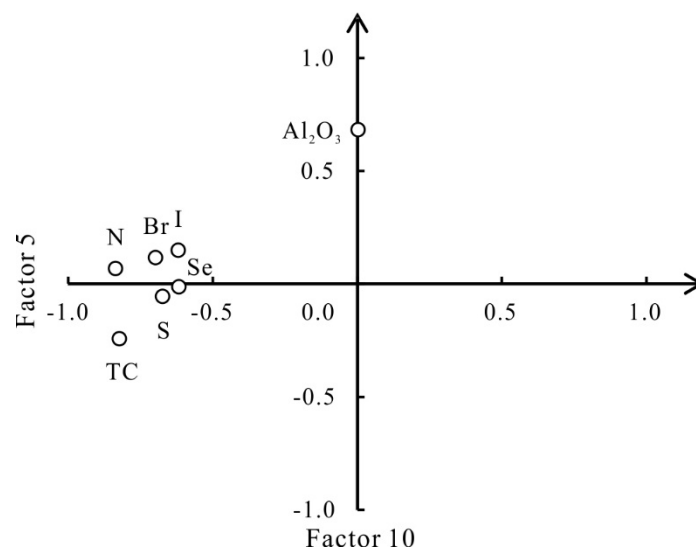


Figure 13. Biplot of F5 (TC–N–S–Br–I–Se association) vs. F10 (Al_2O_3). Circles indicate elements.

The enrichment of Br may be linked to absorption by organic carbon in soils or sediments. The baseline value of I (0.7 mg/kg) is consistent with the average crustal value (0.7 mg/kg). However, the baseline values of F and Cl (271 and 61 mg/kg) were lower than the average crustal values. Br was significantly enriched, whereas Cl was significantly depleted. The natural sources of F in the environment are mainly the weathering of fluoride-bearing minerals [84]. A variety of low-temperature hydrothermal deposits have been developed in areas with high F concentrations (>441 mg/kg). There are sedimentary metamorphic coal mines in the area, and the F concentration in coal, claystone, and other rocks (biotite) in the region is generally high [52,83,85–87]. However, Laos has abundant rainfall and a high-density network of rivers, and F and Cl are easily leached and transported with drainage water [88]. Therefore, the F and Cl concentrations throughout Laos were well below the average crustal values.

The main limitation of this study is that it focused only on the baseline values of the 69 elements and did not consider their spatial distribution. Therefore, it is essential to describe the spatial distribution of the 69 elements in future research.

6. Conclusions

The NGMPL, which covered the entire national territorial area of Laos for sediment sampling and conducted laboratory chemical analyses of 69 elements, has provided high-quality geochemical baseline data. The LGB values of 69 elements obtained from the NGMPL indicate that the medians of Ag, Au, Bi, Cd, Ge, Hg, I, In, Nb, P, Rb, Se, Sn, Ta, Th, U, W, Y, La, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and K₂O are consistent with the average crustal values; As, B, Br, Cs, Hf, Li, N, Pb, Sb, Zr, and SiO₂ are enriched, while Ba, Be, Cl, Co, Cr, Cu, F, Ga, Mn, Mo, Ni, S, Sc, Sr, Ti, Tl, V, Zn, Eu, Al₂O₃, Tot. Fe₂O₃, MgO, CaO, and Na₂O are depleted. Factor analysis shows that the 69 elements can be divided into 12 factors. The origin and enrichment/depletion of elements in different factors related to bedrock (rare earth elements, ferrum-group elements, and the major elements), mineralization (Au, Sb, As, etc.), and farming activities (N, Br, S, and C) are discussed in the context of geological background, mineralization, hypogene processes, and human activities.

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