

Geochemical relations among elements in stream sediment samples from Siojan Prospecting Area, Iran using geostatistical methods

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Abstract Stream sediment samples play an important role in identifying potential areas of metallic and non-metallic mineralization in mineral exploration studies. The relationship of geochemical elements with each other shows how the elements are distributed in the area. Also, by identifying related elements, sampling and targeted chemical analysis can be used in the next stages of exploration. The purpose of this study is to investigate the elements related to the copper element in the Siojan prospecting area, which is located in South-Khorasan province and 30 km northwest of Birjand city of Iran. In Siojan area, 120 stream sediment samples of a 60 square kilometer area were collected to detect geochemical anomalies and were consequently analyzed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) for 45 elements. Preliminary geological studies showed that the studied area has copper mineralization potential, and therefore, copper was selected as the target element in this study. Copper trace elements were identified in the area and the results were used to identify copper mineralized anomalies. For the elemental analysis data, methods of Principal Component Analysis (PCA), Factor Analysis (FA), Hierarchical Cluster Analysis (HCA) and K-Means Clustering were performed to identify the relevant elements and relationships among them. Statistical analysis of the concentration of geochemical elements in the region revealed that copper and cobalt elements were identified as two elements of the same family in terms of geochemical genetics. The average value for copper and cobalt elements in the analyzed samples was 27.2 ppm and 15.5 ppm, respectively. Finally, the relationship between copper and cobalt elements was modeled as an equation using the K-Means Clustering algorithm.

Keywords: Anomaly separation, Clustering, Copper trace elements, PCA, Factor analysis

1 Introduction

Geochemical investigations in mineral exploration are carried out to identify anomalous elements of mineralization (metallic or non-metallic) in the area (Roonwal

2018). In the reconnaissance stage, based on geological and geochemical investigations (sampling), the location of the anomalies of the target element is determined. The importance of this task is to separate the potential areas and barren areas from each other (Gourley 2019, Talapatra 2020). Geochemical relationships between the elements from the stream sediment samples are analyzed by various methods. The most important and practical methods that have been used by researchers in different areas are multifractal analysis, Principal Component Analysis (PCA), clustering, Artificial Neural Network (ANN) and Machine Learning Algorithms (Yin *et al.* 2021, Shirazi 2022, Shirazi *et al.* 2022). Each of the mentioned methods has advantages and disadvantages, which are chosen according to the study purpose (Garcia *et al.* 2020, Mohammadi *et al.* 2018). In the present study, the aim is to identify the elements related to the copper element in the exploratory area of Siojan. For this purpose, Principal Component Analysis (PCA), Factor Analysis (FA), Hierarchical Cluster Analysis (HCA) and K-Means clustering algorithm methods have been chosen. In this way, elemental analysis is performed among all analyzed data from stream sediment samples, and elements related to copper element are identified. The elements detected are called copper trace elements in the area. The Siojan prospecting area is located in South-Khorasan province (East Iran) and 30 km northwest of Birjand city. In terms of structural geology, this area is located in the structural zone of Sistan (Tirrul *et al.* 1983). The studied area has been confirmed in terms of copper mineralization, so that extensive copper oxide and copper sulfide mineralization have been observed in it. Considering that the topic of the current research in this area has not been investigated comprehensively so far, the main objectives of the study are: (1) Descriptive statistics analysis; (2) Determining the geochemical threshold limit of copper element in stream sediment samples of the area; (3) Investigating of geochemical relationships among elements; (4) Separation of mineral elements and rock-forming elements in the area.

2 Methodology

2.1 Study area and geological setting

The study region is located in 32° 58' 23'' N latitude and 58° 56' 7'' E longitude 30 km northwest of Birjand city in South Khorasan of Iran (Figure 1). Siojan area geologically and structurally located in the Philish zone of eastern Iran. Exposed rock units in this area include Phyllis-like sediments (Paleocene-Eocene), limestones,

sedimentary-volcanic facies including marl, tuff and ignimbrite (Eocene) and volcanic rocks (Eocene- Oligocene) including agglomerate, dacite, andesite, and ignimbrite.

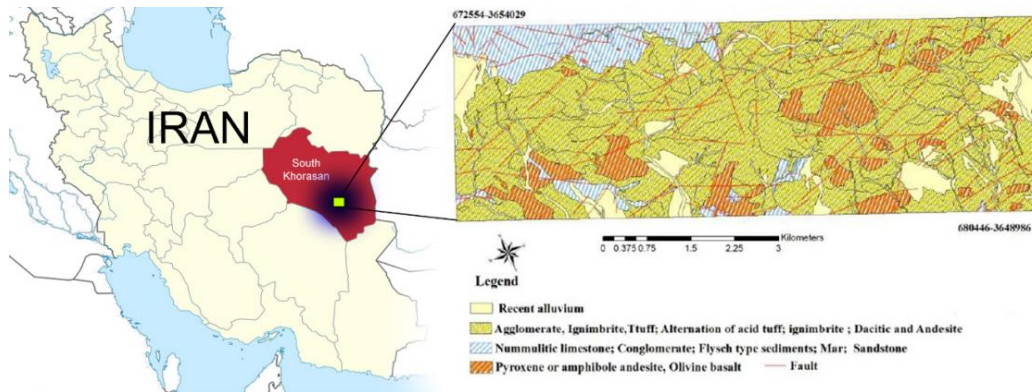


Fig 1. Location and geological map of the area

Alteration zones in this region are seen in relation to Eocene-Oligocene volcanic and around the location of faults and fractures. Alteration zones include argillic alteration in the center and west of the area, siliceous in the center and parts of the south of the area, and carbonation (Malekian *et al.* 2022). In general, the main structural control faults of the region can be divided into two general categories, which include, 1) Northwest-southeast faults that these faults have a greater role in determining the structural status of the region than the second category faults and they often have a non-slip function, and 2) faults with north, northeast-south, southwest, which are more of normal slip slope. In addition to metal mineralization in the study area, small and scattered masses of gypsum can be seen in the south of the study area.

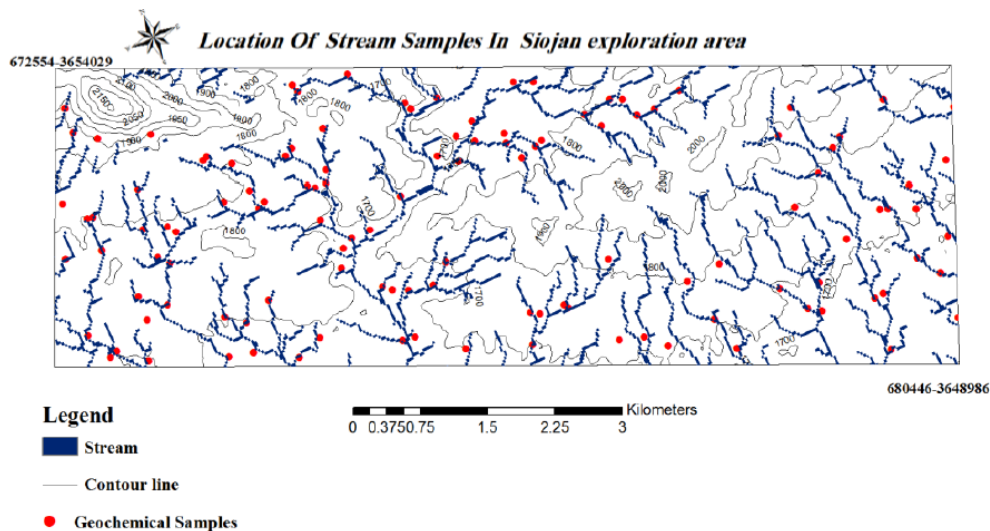


Fig 2. The location of stream sediment samples

2.2 Sampling

Geochemical sampling in Siojan area was prepared through stream sediment sampling method. The initial design of the sampling points was mainly based on determining the center of stream's gravity. For this purpose, a map of the drainage systems (Shirazi *et al.* 2022b) of the study area was generated using topographic maps and aerial images (Figure 2). Stream sediment samples with a particle size of 80 mesh were collected from the study area. The area was covered with 120 stream sediment samples in the form of ICP MS 44 elements analysis (Figure 2) (Shirazi *et al.* 2018a, b, c).

2.3 Technical flow

The most important discussion in the analysis of geochemical data is to determine the background limit for each element in the study area and to separate the background from the anomalies of the relevant element (Carranza 2008). Another important issue in geochemical exploration is the simultaneous study of the elements under study (Alahgholi *et al.* 2018, Shirazy *et al.* 2021 a, b, Zhang *et al.* 2019). One of the most powerful methods in this field is factor analysis as well as principal component analysis. These methods have two advantages (Mohammadi *et al.* 2018): 1) Reducing the dimensions of data, and 2) expressing the existing relationship between different elements. Especially with the large number of elements studied and the large number of samples, the role of factor analysis becomes more apparent, so that it is much easier to understand the variability of the data.

Factor analysis is based on PCA method. This is a technical method for finding a linear combination of the same initial variables to form a new coordinate system. These linear compounds are called principal component analysis and have the following properties (Carranza 2009), 1) most of the variability can be explained by a limited number of new variables, 2) new variables, which are the product of the linear combination of the initial variables, do not show correlation between themselves. Before using this method, it is necessary to pay attention to two points :If the initial variables are not correlated (or have a small correlation coefficient) there is no reason to use this method, because they do not give acceptable results .Factor analysis is performed when the number of initial variables is sufficient .Invoice analysis is performed in four steps:

- 1) Calculation of correlation coefficients,
- 2) Extraction of factors, which includes determining the number and method of calculating factors,
- 3) Applying special transformations on factors, in order to better interpret the relationships between data,
- 4) calculating the score of each factor for each sample.

Clustering is the placement of data in groups where the members of each group are similar to each other on a specific parameter. The similarity between the data within each cluster is maximum and the similarity between the data within different clusters is minimal (Doodran *et al.* 2020; Shirazy *et al.* 2022, Yang *et al.* 2019) .The structure of these clusters or groups can be consistent with the nature of the data or the hidden

structure that lies within the data. Clustering is finding a structure in a set of data that is not categorized. The main reason for using clustering methods is to discover new structures that exist naturally in the data without any prior knowledge of the structure of classes or categories (Nazerian *et al.* 2021, Shirazy *et al.* 2018 d, e, Yu *et al.* 2019). The results of geochemical studies can be useful in future studies in the field of mineral exploration (Beygi *et al.* 2021, Shirazi *et al.* 2018b). For example, in remote sensing surveys and processing of satellite images, they are used in combination to verify the mapping performed (Shirazi *et al.* 2018 b, Shirazy *et al.* 2021 c,d, Shirazy *et al.* 2020 a,b). It is also used in exploratory geophysical studies to integrate information layers and determine the drilling location of boreholes (Khayer *et al.* 2020; Shirazy *et al.* 2021).

3 Results and Discussion

3.1 Descriptive statistical investigation

Due to the polymetallicity of the region, the correlation of elements (Cu, Co, Ag, Sb, Mo, Zn) (Table 1) were identified for analysis. Because our data are not normal, Spearman method was used to determine the correlation.

Table 1: Elements and their correlation with Spearman method.

| | <i>Co</i> | <i>Cu</i> | <i>Mo</i> | <i>Sb</i> | <i>Zn</i> | <i>Ag</i> |
|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| <i>Co</i> | 1 | .586 | .076 | .369 | .204 | .097 |
| <i>Cu</i> | .586 | 1 | .141 | .385 | .419 | .057 |
| <i>Mo</i> | .076 | .141 | 1 | .448 | .131 | .199 |
| <i>Sb</i> | .369 | .385 | .448 | 1 | .255 | .195 |
| <i>Zn</i> | .204 | .419 | .131 | .255 | 1 | .080 |
| <i>Ag</i> | .097 | .057 | .199 | .195 | .080 | 1 |

The statistical results on the data are given in Table 2.

Table 2: Basic statistical results.

| | Min | Max | Sum | Mean | Std. deviation | variance | skewness | kurtosis | | |
|----|-----|-----|------|-------|----------------|----------|----------|----------|--------|-------|
| Ag | 0 | 1 | 42 | 0.35 | 0.202 | 0.041 | 1.298 | 0.221 | 2.659 | 0.438 |
| Co | 9 | 23 | 1860 | 15.50 | 2.961 | 8.770 | -0.013 | 0.221 | -0.080 | 0.438 |
| Cu | 15 | 50 | 3272 | 27.27 | 5.391 | 29.066 | 0.450 | 0.221 | 1.490 | 0.438 |
| Mo | 0 | 3 | 122 | 1.01 | 0.422 | 0.178 | 1.764 | 0.221 | 3.789 | 0.438 |
| Sb | 0 | 7 | 126 | 1.05 | 1.013 | 1.026 | 3.123 | 0.221 | 12.336 | 0.438 |
| Zn | 30 | 68 | 5417 | 45.14 | 8.063 | 65.015 | 0.419 | 0.221 | -0.579 | 0.438 |

3.2 Copper threshold limit investigation

The most important discussion in the analysis of geochemical data is to determine the background threshold for each element in the region and to separate the background threshold from the anomaly of the relevant element (Cheng *et al.* 1996, Khosravi *et al.* 2022, Shirazy *et al.* 2020 c,d, Shirazi *et al.* 2018 a, c, Shirazy *et al.* 2022 a ,b). Using statistical methods, the threshold concentration of geochemical elements is identified. To be used in geochemical modelling and geochemical halo identification. Identifying the concentration threshold of the desired elements is used in mineral processing studies to decide on the economics and how the elements are extracted from the mineral (Doodran *et al.* 2020, Khakmardan *et al.* 2020b, Khakmardan *et al.* 2018).

Anomaly threshold calculator formula:

$$MAD = Median | Xi - Median |$$

According to the above formula: MAD is 3.1 ppm and Median is 27.1 ppm as well as after insertion in the formula:

$$\text{Anomaly} = \text{Median} + 3 \text{ MAD}$$

Our anomalous threshold was estimated to be 33.1 ppm and our anomalous map was created as Figure. Blue indicates anomaly and background is green and other colors indicate anomalous probabilities (Mohammadi *et al.* 2016).

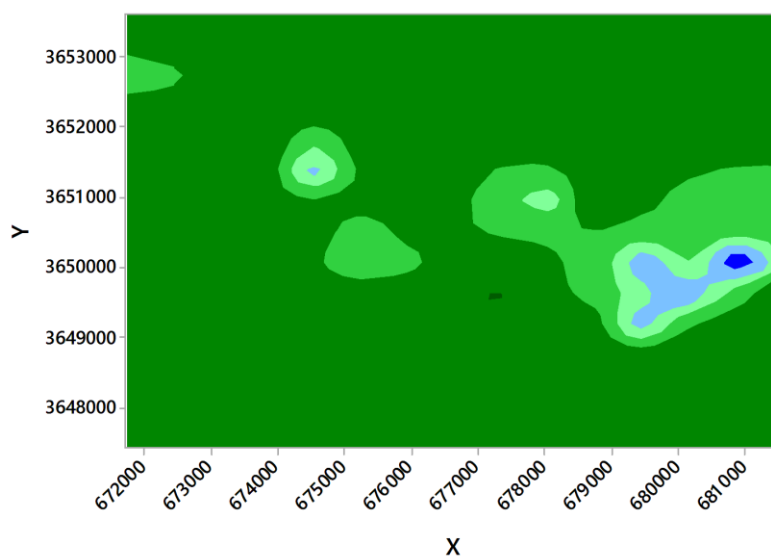


Fig 3. Anomalous areas with blue and green is background
(Coordination in UTM, Zone : 39)

3.3 Principal Component Analysis (PCA)

According to the selected elements, the analysis was performed on the data and the results were announced according to Table 3.

Table 3. Principal component analysis table.

| Variable | PC 1 | PC 2 | PC 3 | PC 4 | PC 5 | PC 6 |
|-------------|--------|--------|--------|--------|--------|--------|
| Cu | 0.513 | -0.371 | -0.048 | 0.04 | 0.296 | 0.712 |
| Co | 0.46 | -0.345 | -0.247 | 0.461 | 0.143 | -0.613 |
| Ag | 0.201 | 0.521 | -0.811 | -0.157 | 0.024 | 0.072 |
| Sb | 0.492 | 0.261 | 0.236 | 0.204 | -0.763 | 0.103 |
| Mo | 0.318 | 0.603 | 0.462 | 0.053 | 0.553 | -0.117 |
| Zn | 0.376 | -0.201 | 0.1 | -0.847 | -0.062 | -0.296 |
| Eigen Value | 2.3101 | 1.1922 | 0.8529 | 0.8148 | 0.4707 | 0.3592 |
| Proportion | 0.385 | 0.199 | 0.142 | 0.136 | 0.078 | 0.6 |
| Cumulative | 0.385 | 0.584 | 0.726 | 0.862 | 0.94 | 1 |

In the Table 3, according to the amount of special values and also the following diagram (Figure 4), the first three components that explain 73% of the behavior were selected.

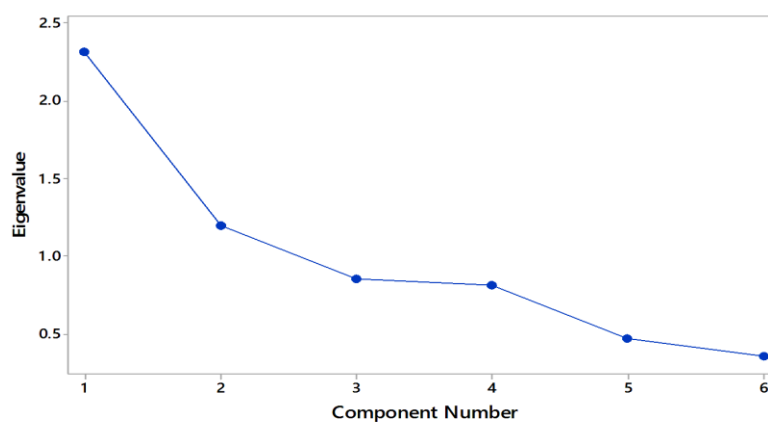


Fig 4. Scree plot in PCA

The outlier data were announced by principal component analysis method with Mahalanobits interval as shown in Figure 5.

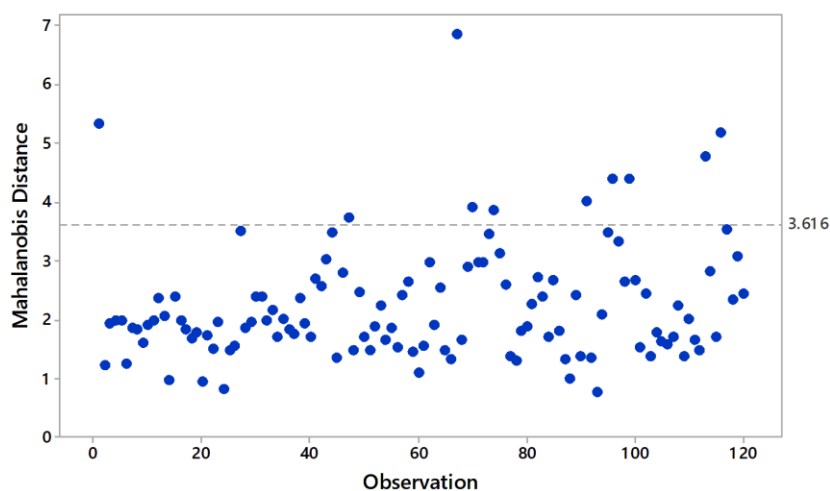


Fig 5. Outlier data detection based on Mahalanobis distance.

Outlier data were replaced with appropriate values. Then the corrected data were used for statistical analysis (Levin 2011, Kim 2013, Kwak and Kim 2017, Alahgholi *et al.* 2018, Aali *et al.* 2022a, b, Shirazy *et al.* 2022a, b, c, d, Shirazi *et al.* 2022a, b).

Based on the principal component analysis (PCA) method, the scores of different elements in each principal component were evaluated. Then the scores of the elements were mapped in each of the first, second and third principal components (PC1, PC2 and PC3). The map obtained from each of the first, second and third principal components are presented in Figures 6, 7 and 8, respectively.

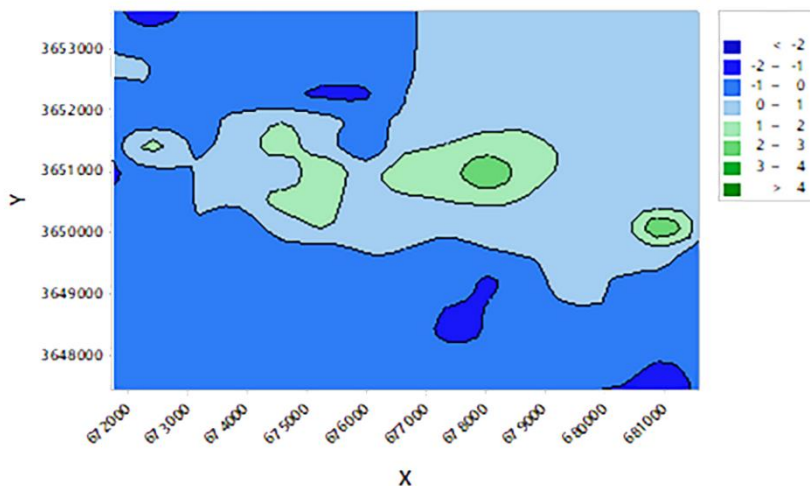


Fig 6. Contour map of principal component analysis method, First component (PC1)
(Coordination in UTM, Zone: 39)

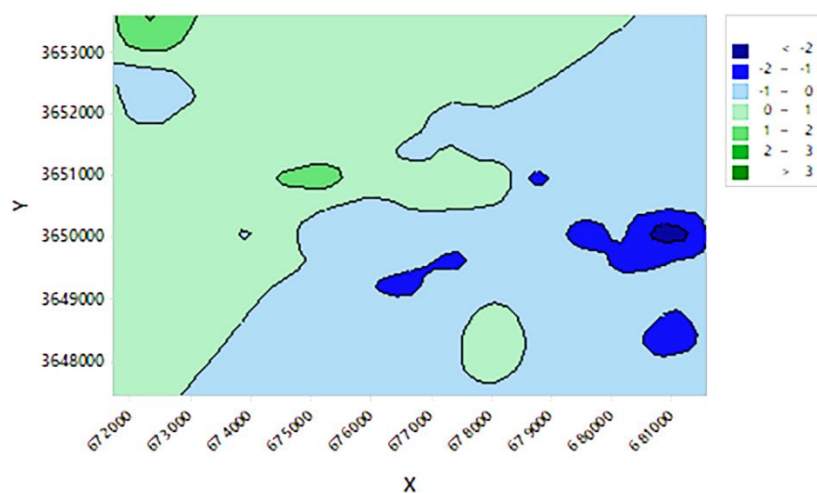


Fig 7. Contour map of principal component analysis method, second component (PC2)
(Coordination in UTM, Zone: 39)

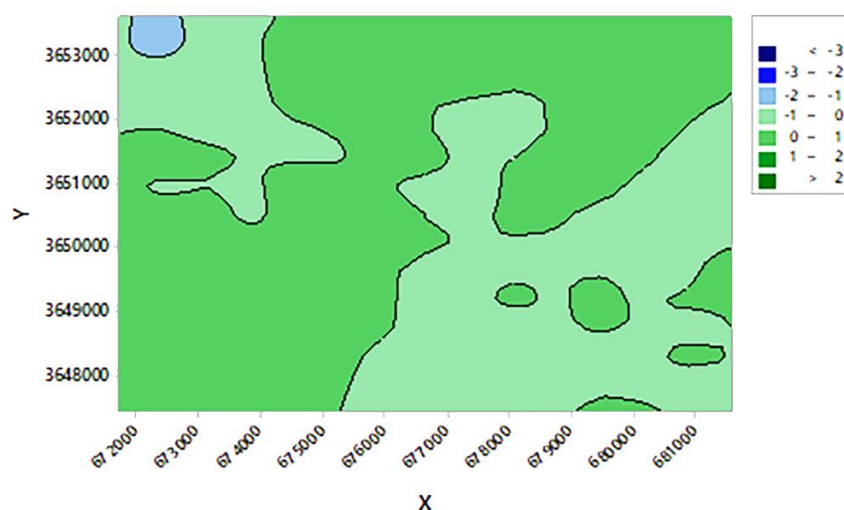


Fig 8. Contour map of principal component analysis method, third component (PC3)
(Coordination in UTM, Zone: 39)

The studied elements were clustered using principal component analysis (PCA) method. For this purpose, the loads related to each of the elements in the first and second main components were plotted against each other. Elements close to each other were grouped together. Based on this clustering, 3 different group classes were obtained. Silver and molybdenum elements group, zinc, cobalt and copper elements group and antimony single element group (Figure 9).

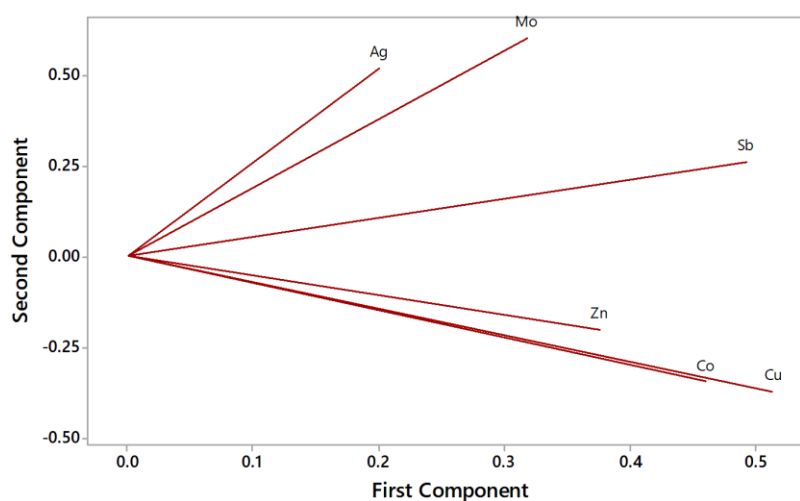


Fig 9. Clustering by PCA method using components one and two.

3.4 Factor Analysis (FA)

Factor analysis is a multivariate statistical method that establishes a special relationship between a large set of seemingly unrelated variables under a hypothetical model. In factor analysis, a large number of variables are expressed in terms of a small number of dimensions or structures; this structure is called a factor. In methods based on eigenvectors using eigenvalues and eigenvectors, directions with maximum variability are identified. In order to analyze the geochemical data of stream sediments, the rotary type factor analysis method with variomex rotation was used (Reimann *et al.* 2005, Shirazy *et al.* 2018a, b, c, Shirazy *et al.* 2019, Shirazi *et al.* 2021, Hedayat *et al.* 2022, Shirazy *et al.* 2022c).

In Table 4, by selecting 3 factors and also the method of maximum likelihood, the numbers for each factor in each element are shown in Figures 10 to 12.

Table 4. Factor analysis table.

| Variable | Factor 1 | Factor 2 | Factor 3 | Communality |
|----------|----------|----------|----------|-------------|
| Cu | 0.63 | -0.35 | 0.398 | 0.678 |
| Co | 0.979 | 0.057 | -0.25 | 0.962 |
| Ag | 0.106 | -0.213 | -0.177 | 0.088 |
| Sb | 0.405 | -0.574 | -0.179 | 0.526 |
| Mo | 0.104 | -0.599 | -0.344 | 0.488 |
| Zn | 0.239 | -0.383 | 0.338 | 0.318 |
| Variance | 1.5979 | 1.0056 | 0.4555 | 3.0589 |
| % Var | 0.266 | 0.168 | 0.076 | 0.51 |

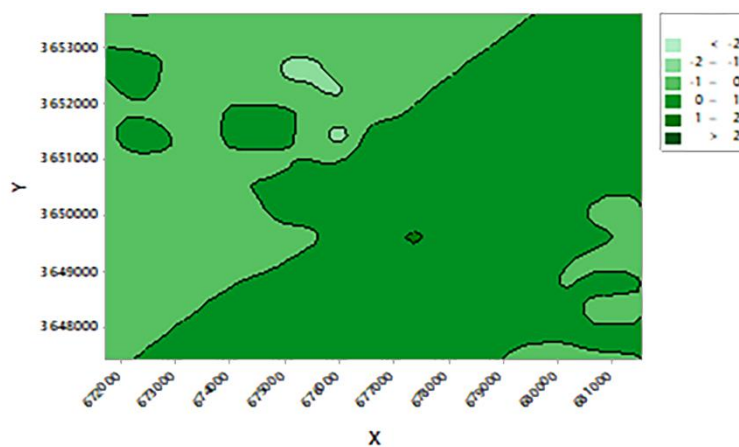


Fig 10. Contour map of factor one (Coordination in UTM, Zone: 39)

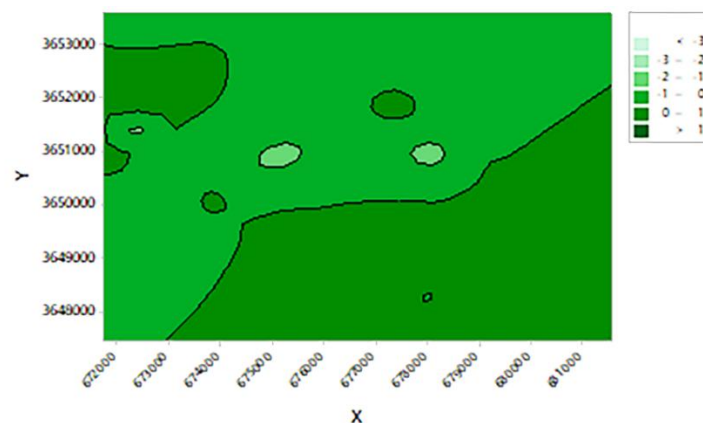


Fig 11. Contour map of factor two (Coordination in UTM, Zone: 39).

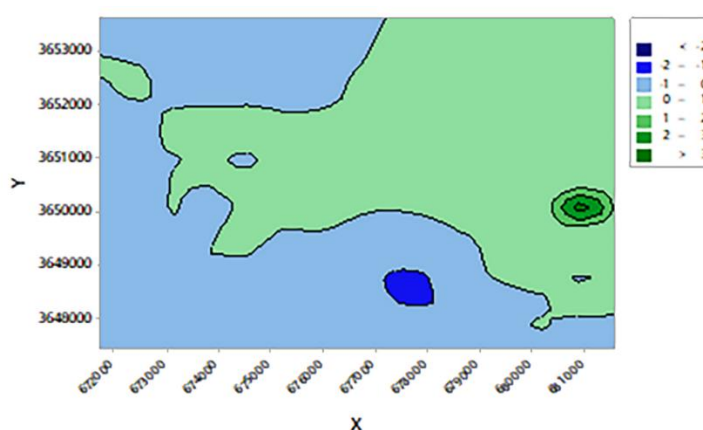


Fig 12. Contour map of factor three (Coordination in UTM, Zone: 39)

According to the loads in the first two factors, Figure 13 can be drawn.

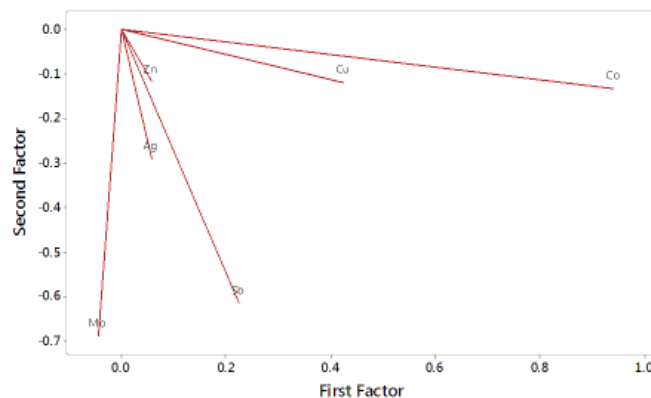


Fig 13. Factor classification according to factor one and two.

Or in the case of factors one and two, it is shown schematically in Figure 14.

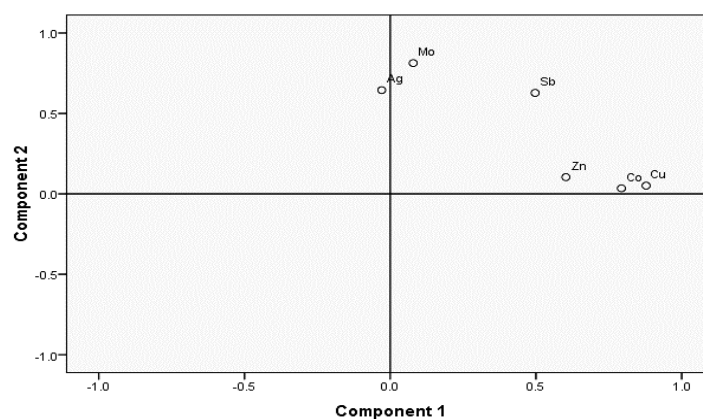


Fig 14. Factor classification according to factor one and two schematically.

Based on the obtained results of factor analysis method, another grouping of the studied elements can be presented. This grouping includes cobalt, copper and zinc groups, silver and molybdenum groups and single element antimony group.

3.5 Hierarchical Cluster Analysis (HCA)

In data mining and statistics, "hierarchical clustering" is a method that performs the act of categorizing and grouping observations and data in a hierarchical manner. The point that distinguishes this method from other clustering methods is the sequence and top-down (or bottom-up) view that exists in this technique (Khakmardan *et al.* 2020a; Khayer *et al.* 2021; Madani *et al.* 2021; Nazerian, Catania, *et al.* 2022; Nazerian,

Shirazy, *et al.* 2022). For clustering, the hierarchical distance clustering method was used and the results were presented according to Table 5 and the dendrogram in Figure 15.

Table 5: Hierarchical clustering parameters.

| Agglomeration Schedule | | | | | | |
|------------------------|------------------|-----------|--------------|-----------------------------|-----------|------------|
| Stage | Cluster Combined | | Coefficients | Stage Cluster First Appears | | Next Stage |
| | Cluster 1 | Cluster 2 | | Cluster 1 | Cluster 2 | |
| 1 | 3 | 5 | 74.721 | 0 | 0 | 2 |
| 2 | 3 | 4 | 136.962 | 1 | 0 | 4 |
| 3 | 1 | 2 | 18884.820 | 0 | 0 | 4 |
| 4 | 1 | 3 | 57148.272 | 3 | 2 | 5 |
| 5 | 1 | 6 | 177736.097 | 4 | 0 | 0 |

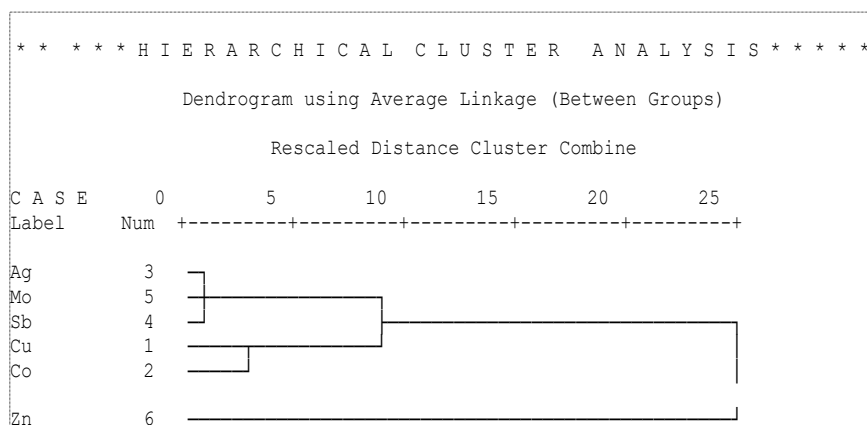
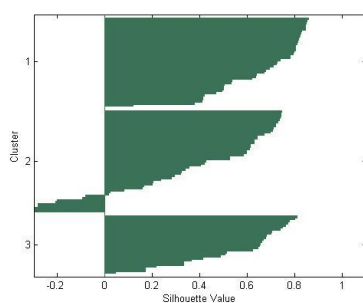


Fig 15. Final hierarchical clustering dendrogram.

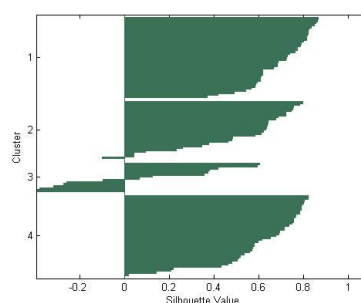
Geochemical elements can be grouped according to the results of hierarchical analysis. In this grouping, silver, molybdenum and antimony elements are in one group, copper and cobalt elements in one group and zinc element as a single element in a separate group.

3.6 K-Means Clustering Analysis

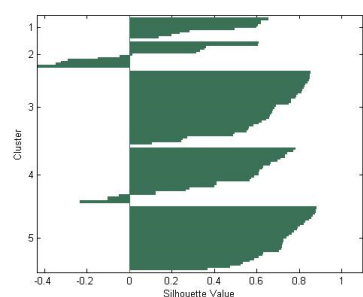
As it was observed, according to the results of statistical analysis, copper and cobalt elements are inherently geochemically related to each other. Therefore, the geochemical behavior of these two elements in relation to each other (as an exploratory key in the study region) (Shirazy *et al.* 2019), is important. In order to investigate the geochemical behavior of Cu and Co elements, K-Means clustering method was used (Khayer *et al.* 2020, Shirazy *et al.* 2022c).



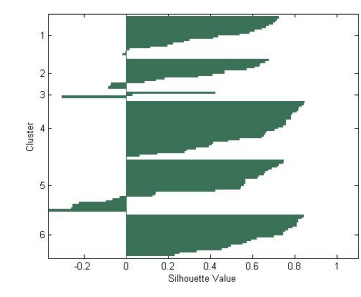
Classification of 3 classes, with an average value of 0.5237



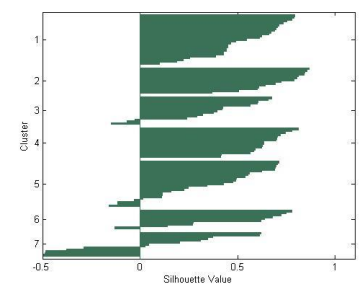
Classification of 4 classes, with an average value of 0.5556



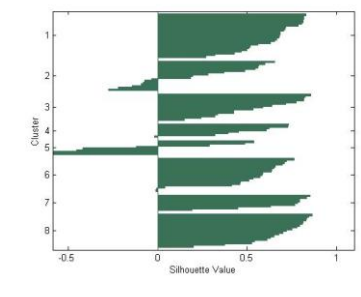
Classification of 5 classes, with an average value of 0.5468



Classification of 6 classes, with an average value of 0.4677



Classification of 7 classes, with an average value of 0.4471



Classification of 8 classes, with an average value of 0.4975

Fig 16. Profile images obtained from the classification of Cu and Co elements along with the mean values of the utility function.

Initially, the data set was categorized into different classes using the K-Means algorithm. Then the amount of utility function for each classification was calculated. As shown in Figure 17, the image of the silhouettes is presented along with the mean value of the utility function.

Based on the results of geochemical data classification of copper and cobalt elements, classification of 4 classes with the highest value of the utility function was selected as the optimal clustering (Ahmadi *et al.* 2022, Shirazy and Hezarkhani 2018,

Shirazy *et al.* 2022; Shirazy *et al.* 2021). The graph of the values of the utility function for classifying 3 classes to 20 classes is presented in Figure 17.

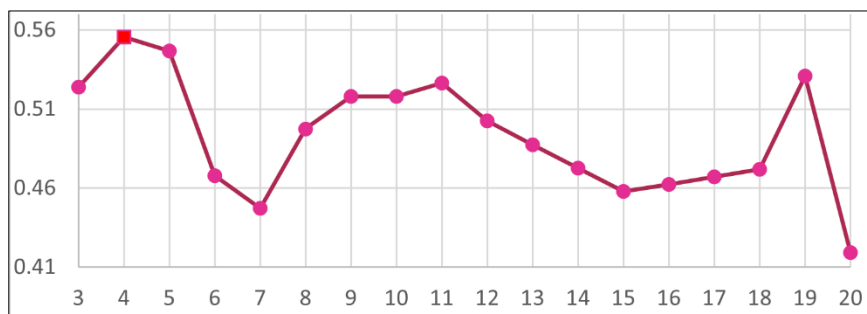


Fig 17. The graph of the values of the utility function (silhouette) for classifying 3 classes to 20 classes.

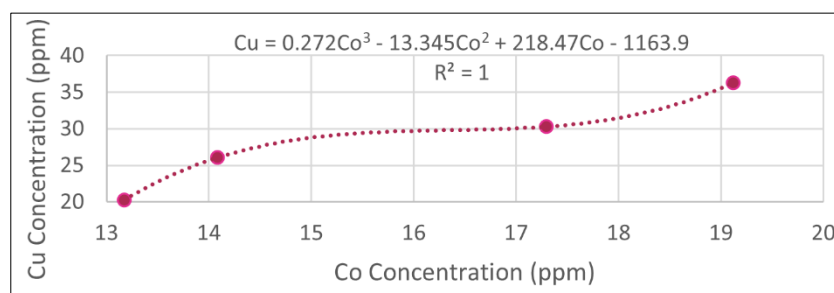


Fig 18. Geochemical behavior of copper (Cu) vs. cobalt (Co) elements.

Cu and Co concentrations were calculated in the centers of 4-class classification based on K-Means algorithm. Using regression method, the values of the centers of the clusters were plotted. The diagram shown in Figure 18 is actually the geochemical behavior of copper and cobalt elements in samples of stream sediments in the Siojan region. As can be seen in the geochemical behavior diagram of the elements copper and cobalt, the concentrations of these two elements are directly related to each other. Thus, with increasing Cu concentration, Co concentration also increases. This increase in value is calculated as a quadratic curve. The third-degree equation of geochemical behavior of copper and cobalt concentrations is:

$$Cu = 0.272Co^3 - 13.345Co^2 + 218.47Co - 1163.9$$

4 Conclusions

Siojan exploration region is located in South Khorasan province of Iran with the mineralization potential of copper element. Copper mineralization has occurred in this area. Therefore, it was used as a target for studies to identify geochemical anomalies.

120 geochemical samples of stream sediments were collected according to the designed sampling map. After preparing the samples, they were analyzed by ICP-MS method. From each sample, 44 elements were obtained as a result of analysis. Data were prepared using statistical methods. Then, in order to identify the relationships of geochemical elements in the region, the methods of principal component analysis, factor analysis and hierarchical analysis were used.

The grouping performed on the geochemical elements by the methods of principal component analysis and factor analysis found similar results. Based on these results, three groupings of geochemical elements of the region were presented. Cu, Co and Zn group, Ag and Mo group and single element Sb group. According to the same result, the methods of principal component analysis and factor analysis in the grouping of geochemical elements in the region, in order to increase the accuracy of research and identify elements related to the element, hierarchical analysis was used. Based on this method, geochemical elements showed different groupings. This grouping also had similarities with the grouping obtained from the results of other methods. According to this grouping, the elements Ag, Mo and Sb in one group, Cu and Co elements in one group and Zn as a single element in a separate group. Due to the fact that the target element in this study was copper, based on the results of statistical studies, the genetic family of this element in Siojan region is cobalt. Although the element zinc was also included in the two groups, which was part of the copper group family by the method of principal component analysis and factor analysis, but it cannot be mentioned as an intrinsic relationship in the region. Another reason is the inherent and permanent correlation between zinc and lead, which was not achieved in statistical analysis.

Geochemical behavioral studies of Cu and Co elements based on K-Means algorithm show that these two elements have direct behavior with each other. Behavioral changes in the concentration of elements relative to each other were calculated as a quadratic equation. Using this equation, the concentration of each element can be calculated based on the other element. The calculated equation is

$$\text{Cu} = 0.272\text{Co}^3 - 13.345\text{Co}^2 + 218.47\text{Co} - 1163.9.$$

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