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Compositional variability of minerals in outcrops and boreholes within the informal Bistra Formation, Medvednica Mt., Croatia

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Abstract

This study investigates the compositional variability of minerals in the informal Bistra Formation at Medvednica Mountain, Croatia, focusing on Quaternary sediments from outcrops and boreholes. Employing modal analysis and compositional data (CoDa) statistical methodologies, the research aims to classify samples and establish associations of minerals to enhance the understanding of sediment differentiation. The analysis reveals significant variations in mineral compositions influenced by local depositional environments and weathering of specific geological formations. Key findings include the identification of two distinct depositional environments: high-energy riverine settings, which concentrate heavier minerals, and low-energy lakes or swamps, where lighter minerals accumulate. The utilization of principal component and cluster analysis further elucidates the relationships among minerals. This approach shows the usefulness of CoDa analysis in geosciences, providing deeper insights into the provenance and deposition dynamics of Quaternary sediments.

Keywords: Bistra Formation; Medvednica Mountain; modal analysis; compositional data analysis; classification

1. Introduction

The informal Bistra Formation is a lithostratigraphic unit encompassing Quaternary sediments around Medvednica Mountain. The Bistra Formation was deposited in an alluvial environment in the areas of sheet flows, breakthrough channels, longitudinal streams, and floodplains (**Avanić, 1997**; **Grizelj et al., 2017**). Similar Quaternary deposits, characterized by short transport and local provenance, can be found throughout the Pannonian Basin (**Grizelj et al., 2017**; **Nenadić et al., 2016**; **Mencin Gale et al., 2019**; **Zámolyi et al., 2017**).

These sediments were further subdivided chronologically using dating methods, and lithologically into clays, silts, sand, and gravel. However, they were not subdivided based on mineralogical composition. Although the provenance is local, the mineralogical composition can vary based on factors such as the type of source rocks and the intensity of weathering (**Peh et al., 1998**; **Novaković et al., 2018**). Different types of source rocks contribute distinct minerals to the sediments, while weathering processes can alter the original minerals and create new ones.
The aim of this study was to classify samples into groups and establish associations between minerals with minimal loss of information. The purpose of this study was to improve the differentiation of Quaternary sediments, which could be further complemented by dating, granulometric and chemical composition, and/or other methods.

2. Geographical and geological setting

The study area encompasses the south-southeastern foothills of Medvednica Mountain, extending into parts of the lower city of Zagreb (**Figure 1**). Medvednica Mountain, rising to an elevation of 1,035 meters at its peak, is situated in northwestern Croatia, just north of the city of Zagreb.

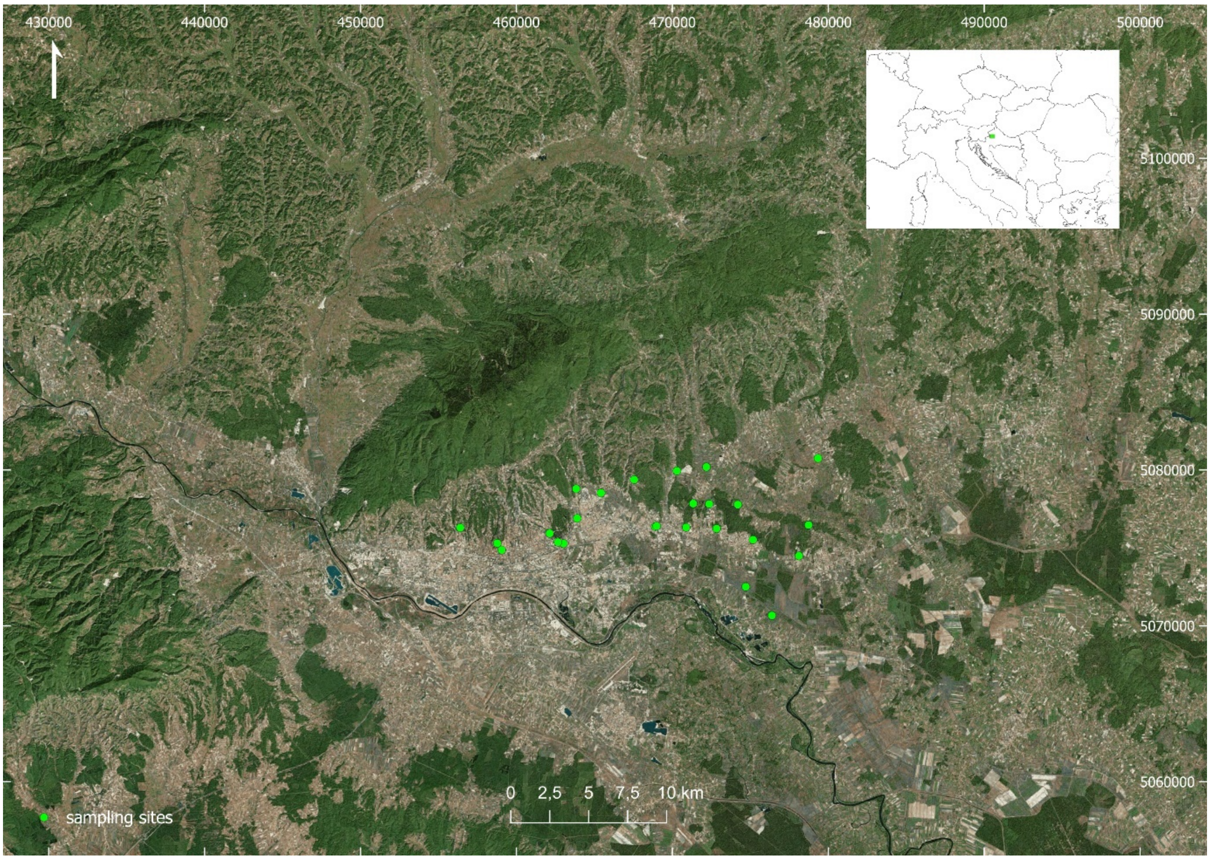

Figure 1: Study area (HTRS96 Projected coordinate system)

The geological characteristics of the study area, including sampling locations, are shown in **Figure 2**. The south-southeastern foothills of Medvednica Mountain are part of the southwestern Pannonian Basin System (**Pavelić and Kovačić, 2018**). The core of Medvednica Mountain consists mainly of metamorphic rocks, such as metapelites, metapsammites, slates, quartzites, marbles, and schists, originating from the Paleozoic and less frequently the Mesozoic eras (**Belak et al., 2022; Lugović et al., 2006**). Additionally, ophiolite mélange fragments (**Slovenec and Lugović, 2009**) and Mesozoic limestones and dolomites are present. Miocene sediments overlie the crystalline basement, consisting of various marine, brackish, and freshwater clastites. Pliocene and Quaternary sediments include clays, silts, sands, and gravels deposited in freshwater lakes, swamps, and rivers (**Basch, 1983a, 1983b**; **Vrsaljko et al., 2006**; **Kovačić and Grizelj, 2006**; **Ćorić et al., 2009**; **Brlek et al., 2016**; **Grizelj, 2017**).

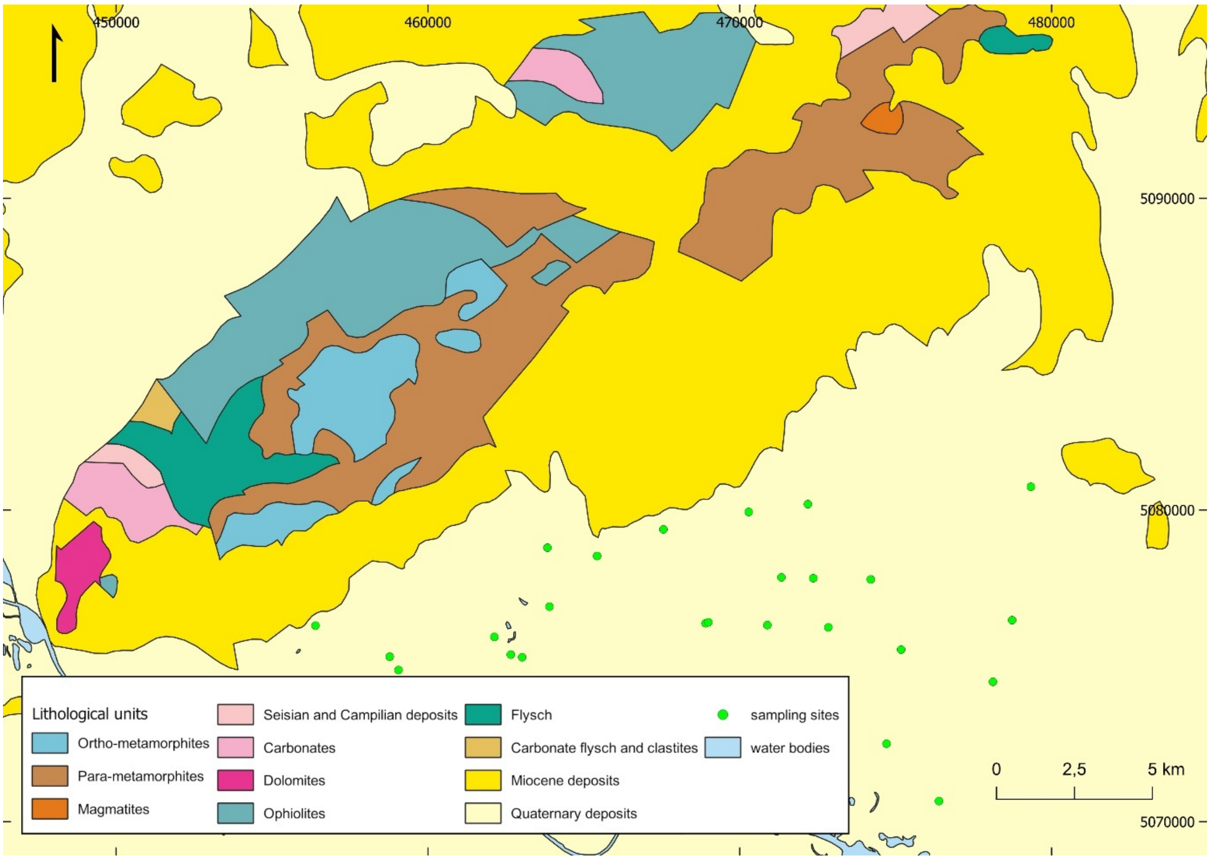

Figure 2: Geological map of the study area at a scale of 1:300,000 (simplified after **the Croatian geological survey** (**2009**); HTRS96 Projected coordinate system)

3. Materials and Methods

Data set consists of 45 samples collected from boreholes and outcrops analysed for the purpose of various projects including Geological map of the Republic of Croatia. The mineral composition of the samples at different depths was determined through modal analysis of fractions between 0.09 and 0.045mm. Calcite in the samples was dissolved using cold hydrochloric acid, and the remainder was separated into heavy and light mineral fractions using bromoform liquid. Mineral counting was performed on 300-400 grains using a microscope. The complete data set consisted of 14 variables in terms of minerals and 45 samples (**Table 1**). However, modal analysis resulted with 68 non-detects (10%). Thus, the data set represents compositional count data, consisting of non-negative integer values representing counts of different minerals.

Table 1: Counts of mineral grains (ZC – zero counts)

Sample	**Opaque minerals**	**Chlorite**	**Tourmaline**	**Zircon**	**Rutile**	**Amphibole**	**Epidote-Zoisite-Clinozoisite**	**Garnet**	**Kyanite**	**Staurolite**	**Quartz**	**K-feldspar**	**Rock fragments**	**Muscovite**
1	6	ZC	1	ZC	1	7	290	ZC	1	ZC	192	61	23	ZC
2	12	ZC	2	1	1	13	264	7	2	1	110	29	176	1
3	4	1	1	1	1	5	301	1	ZC	1	117	147	23	ZC
4	4	ZC	ZC	ZC	ZC	7	284	ZC	ZC	2	142	117	50	1
5	3	1	1	ZC	ZC	6	290	ZC	ZC	ZC	88	176	35	ZC
6	5	ZC	1	ZC	2	2	298	1	3	ZC	198	84	23	1
7	88	1	3	9	2	1	188	6	1	3	218	30	53	2
8	69	1	1	4	6	15	192	12	1	3	182	13	98	3
9	80	4	7	2	12	6	140	25	5	24	254	17	25	12
10	22	ZC	13	5	4	1	228	19	1	7	263	13	29	1
11	23	3	2	1	3	ZC	259	10	1	3	219	7	73	1
12	25	ZC	2	ZC	2	1	278	1	ZC	ZC	211	20	72	1
13	19	1	3	1	1	4	257	8	1	10	228	22	57	1
14	76	2	7	3	2	14	155	37	1	13	202	18	84	1
15	39	1	3	1	2	5	241	17	2	5	188	33	84	ZC
16	17	12	ZC	3	2	8	270	3	ZC	1	119	17	163	1
17	28	ZC	2	1	ZC	4	259	13	1	3	233	22	53	1
18	11	ZC	1	1	ZC	7	284	2	1	3	235	38	42	1
19	102	7	25	3	14	8	100	3	7	20	263	35	13	1
20	77	1	28	1	21	2	132	3	8	23	240	37	16	9
21	202	3	5	4	4	6	84	5	ZC	1	181	18	104	ZC
22	210	2	22	3	11	9	103	5	1	7	219	27	51	1
23	83	5	21	1	4	5	147	31	4	10	244	32	39	5
24	163	1	14	15	19	ZC	86	10	2	3	229	44	26	1
25	89	3	13	7	33	4	123	11	10	16	226	60	19	2
26	58	3	31	3	19	4	175	2	7	9	239	36	14	15
27	41	5	5	2	4	22	212	5	3	12	166	36	107	1
28	97	ZC	2	ZC	5	10	194	ZC	ZC	1	172	62	75	ZC
29	80	ZC	4	2	4	ZC	204	1	1	5	167	33	101	2
30	20	2	2	ZC	ZC	24	252	2	ZC	2	165	64	73	4
31	13	ZC	1	1	3	2	282	1	ZC	1	207	88	13	1
32	71	4	15	12	12	1	129	ZC	100	18	180	32	31	52
33	48	ZC	23	ZC	13	1	167	ZC	24	25	202	41	39	1
34	87	ZC	13	4	11	2	130	5	13	39	256	30	14	2
35	70	1	24	2	16	2	111	3	6	69	241	32	18	2
36	115	7	37	3	25	3	67	2	6	39	275	15	14	3
37	43	11	23	7	8	10	139	26	2	28	239	37	9	12
38	137	ZC	6	2	15	7	101	31	1	5	220	22	65	1
39	23	9	1	3	1	54	215	4	1	1	247	25	17	15
40	151	16	2	3	20	14	65	21	1	6	203	9	85	3

41	8	ZC	2	ZC	2	32	249	6	ZC	1	183	79	40	1
42	15	2	1	ZC	1	15	271	ZC	1	1	156	65	81	1
43	77	51	15	ZC	21	3	91	13	6	26	230	45	9	10
44	56	ZC	25	4	10	12	135	7	2	12	232	31	13	19
45	6	ZC	1	ZC	1	2	301	ZC	1	3	138	163	1	ZC

Traditional statistical methods may not be appropriate due to the data's constrained nature (**Pearson, 1987**; **Pawlowsky-Glahn and Egozcue, 2006**). Therefore, compositional data (CoDa) methodology was applied (**Aitchison, 1982**). CoDa methodology deals with positive values where multiple parts contribute to a whole. However, zeros in CoDa pose a significant challenge, either samples or variables containing zeros need to be removed which results with information loss, either they need to be addressed in some way. Zeros can occurr due to rounding, they can represent essential non-occurrence, or they can be a result of a counting process itself (**Martín-Fernández et al., 2003**).

One effective approach for handling zeros in compositional count data is the Bayesian-multiplicative method, specifically designed to handle count zeros, which often result from an insufficient number of trials rather than the absence of a category (**Martín-Fernández et al., 2015**). This method minimizes distortion in the covariance structure and transforms the data using log-ratio transformations to stabilize variances and make the distribution more symmetric, helping to meet the normality assumptions required for the parametric Bayesian approach.

Following replacement of count zeros, data set was described with normalized (**Egozcue et al., 2013**) variation matrix (**Aitchison, 1986**) containing the variances of all pairwise logratios, and a compositional (e.g. **Filzmoser et al., 2018**) biplot (**Gabriel, 1971**). Finally, groups of components and samples were determined according to the principal component analysis (PCA) loadings and scores, respectively, but also by the means of cluster analysis for compositional data (e.g. **Filzmoser et al., 2018**).

Statistical analysis, including production of diagrams was done in R 4.2.1 (**R Core Team, 2022**). Packages used in analysis were *compositions* (**van den Boogaart et al., 2024**), *robCompositions* (**Templ et al., 2023**) and *zCompositions* (**Palarea-Albaladejo and Martín-Fernández, 2024**). Maps were produced in QGIS 3.34.1 (**QGIS Development Team, 2023**).

4. Results and discussion

Figure 3 illustrates the patterns of zero counts across samples and minerals. Chlorite, zircon, and kyanite showed the highest occurrences of zero counts. Chlorite was frequently absent across the samples, often missing in conjunction with rutile. Since the rutile is indicative of high-grade metamorphism, the absence of both minerals in these samples could suggest a lack of material contribution from retrograde altered high-grade metamorphic rocks present at the Medvednica Mountain (**Belak et al., 2022**). Kyanite and muscovite were typically absent together, while amphibole was commonly found to be missing on its own. Kyanite and muscovite could be associated with metapelitic rocks where kyanite forms by metamorphose of clay sediments (e.g. **Slovenec and Bermanec, 2003**), thus, their absence in samples could indicate low input of weathered material from areas containing such rocks.

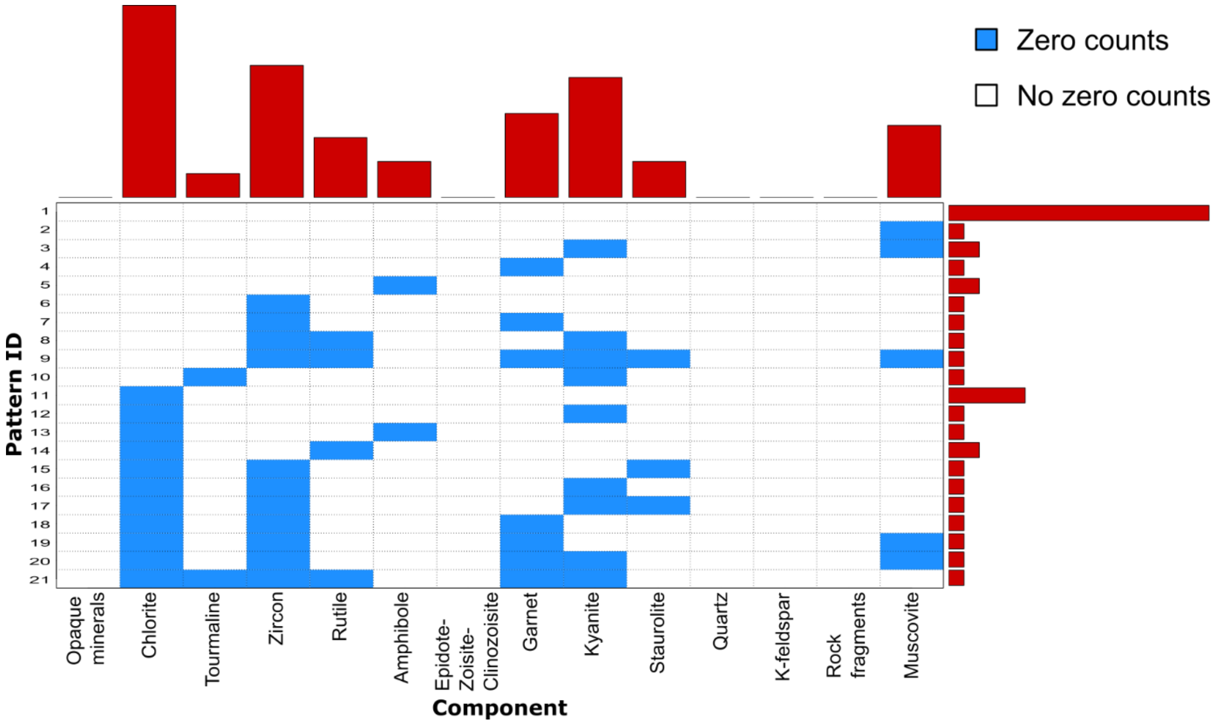

Figure 3: Distribution of zero counts for mineral grains across samples. Zero counts are displayed at the top of each column, and the counts of patterns are indicated on the right side of the table.

The variation matrix (**Table 2**) shows normalized values that indicate the degree of proportionality between pairs of minerals (values closer to zero indicate better proportionality). Low values among rutile, tourmaline, kyanite, staurolite, zircon, and muscovite suggest good proportionality among these minerals, likely indicating a similar source of metamorphic origin. Similarly, low values among epidote-zoisite-clinozoisite, quartz, and K-feldspar indicate their common source. Amphibole shows the highest values in the variation matrix, possibly pointing to its unique source or multiple sources, which affects its variance with other components. Relatively high values of chlorite with all minerals in the variation matrix could indicate its hydrothermal origin, which is widespread and not specifically tied to any single rock type.

Table 2: Variation matrix showing log-ratio variances of the corresponding two parts of the composition (variable in column stands for numerator, while variable in row stands for denominator)

	Chlorite	Tourmaline	Zircon	Rutile	Amphibole	Ep-Zo-Czo	Garnet	Kyanite	Staurolite	Quartz	K-feldspar	Rock fragments	Muscovite
Opaque minerals	0.93	0.48	0.51	0.32	1.48	1.24	0.80	0.83	0.67	0.54	1.45	1.04	0.88
Chlorite		1.20	1.01	0.96	1.07	1.17	1.10	1.11	1.15	0.77	1.35	1.32	0.79
Tourmaline			0.81	0.32	1.89	1.41	1.15	0.49	0.31	0.69	1.40	1.73	0.81
Zircon				0.68	1.48	1.07	0.81	0.94	0.93	0.59	1.36	1.15	0.82
Rutile					1.69	1.27	1.11	0.49	0.48	0.57	1.26	1.48	0.75
Amphibole						0.71	1.24	1.75	1.90	0.73	0.84	0.88	1.25
Ep-Zo-Czo							1.22	1.03	1.44	0.20	0.23	0.51	0.99
Garnet								1.55	1.11	0.75	1.71	1.14	1.23
Kyanite									0.47	0.61	1.02	1.53	0.62
Staurolite										0.77	1.53	1.87	0.83
Quartz											0.39	0.58	0.59
K-feldspar												1.02	1.15
Rock fragments													1.48

Figure 4 shows the form and covariance compositional biplot, respectively. Representation of samples is better in form, while variables in covariance biplot. The first two principal components (PC1 and PC2) explain 55% of the variability (37% and 18% respectively). Tourmaline, rutile, and staurolite show strong association and significant positive loadings on the first principal component (PC1+). Kyanite, zircon, and opaque minerals also exhibit positive loadings on PC1. Since all these minerals represent the heavy fraction and are relatively resistant mineral phases, it is likely that PC1+ represents a placer-like depositional environments, where heavier and more durable minerals are concentrated due to high-energy conditions such as those found in riverine settings. Conversely, epidote-zoisite-clinozoisite, K-feldspar, and quartz show strong association and significant negative loadings on PC1 (PC1-). All these components, except for amphibole and epidote-zoisite-clinozoisite, represent the light mineral fraction. Including rock fragments which were mostly made of greenschists (**Grizelj, 2017**), indicating they were composed of mostly light mineral fractions. Some amphiboles have densities closer to the boundary between what is typically considered heavy and light which could explain why are amphiboles grouped with other minerals of lighter fraction. Epidote-zoisite-clinozoisite has a similar density to tourmaline and is lighter than all other minerals significantly associated with PC1+. Accordingly, PC1- could represent depositional environments of low energy, such as lakes and swamps, where lighter mineral fraction could accumulate away from high-energy erosive processes.

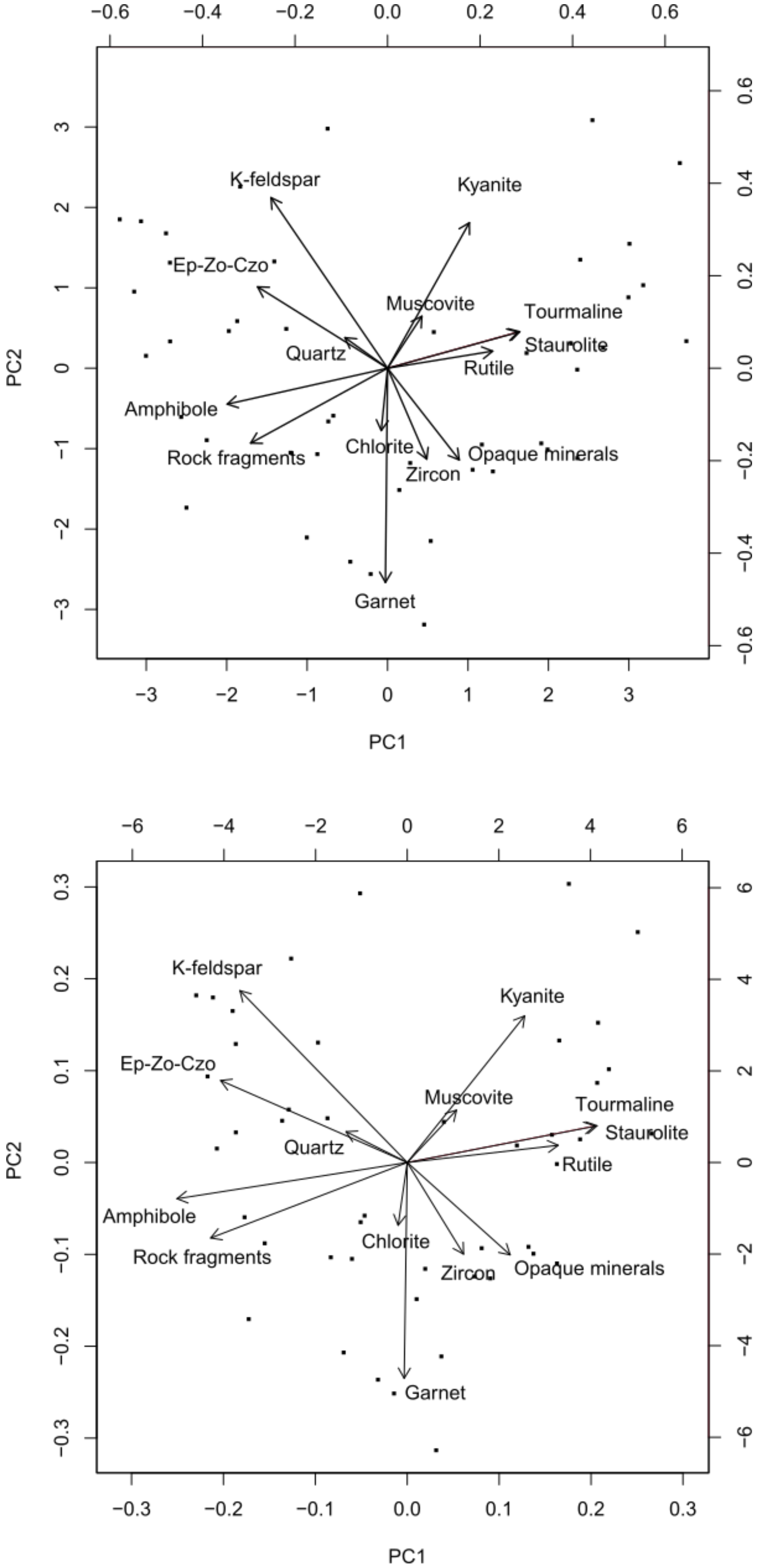

Figure 4: Form (top) and covariance (bottom) compositional biplots

The distribution of loadings on PC1 suggests it captures local variations in depositional environments, correlating high loadings with high-energy environments (e.g., rivers) and low loadings with low-energy environments (e.g., lakes and swamps). PC2, explaining only 18% of the variability in the dataset, indicates a need for more detailed investigation. However, garnet, chlorite, zircon, and opaque minerals show the lowest negative loadings on PC2 (PC2-), while kyanite, muscovite, and K-feldspar show positive loadings on PC2 (PC2+). Possibly, PC2 differentiates between weathering processes of different rock types present on Medvednica Mountain. For example, PC2+ could represent the weathering process of metamorphic rocks, while PC2- could represent a redepositional process of the Miocene clastites surrounding the crystalline core of Medvednica Mountain.

Cluster analysis yielded results similar to those of PCA (**Figure 5**). Two main groups of samples are visible. The second group (represented by pluses and triangles in **Figure 5** on the right), which consists of heavy minerals (except for muscovite and chlorite), can be divided into two subgroups. This division likely points to two different sources of weathering: metamorphic rocks and the Miocene clastites, which are predominantly present in the southeastern part of Medvednica Mountain. The first group (circles in **Figure 5** on the right) likely contains minerals found in the majority of rocks on Medvednica Mountain but are deposited in locally calmer environments, as they represent mostly the light mineral fraction.

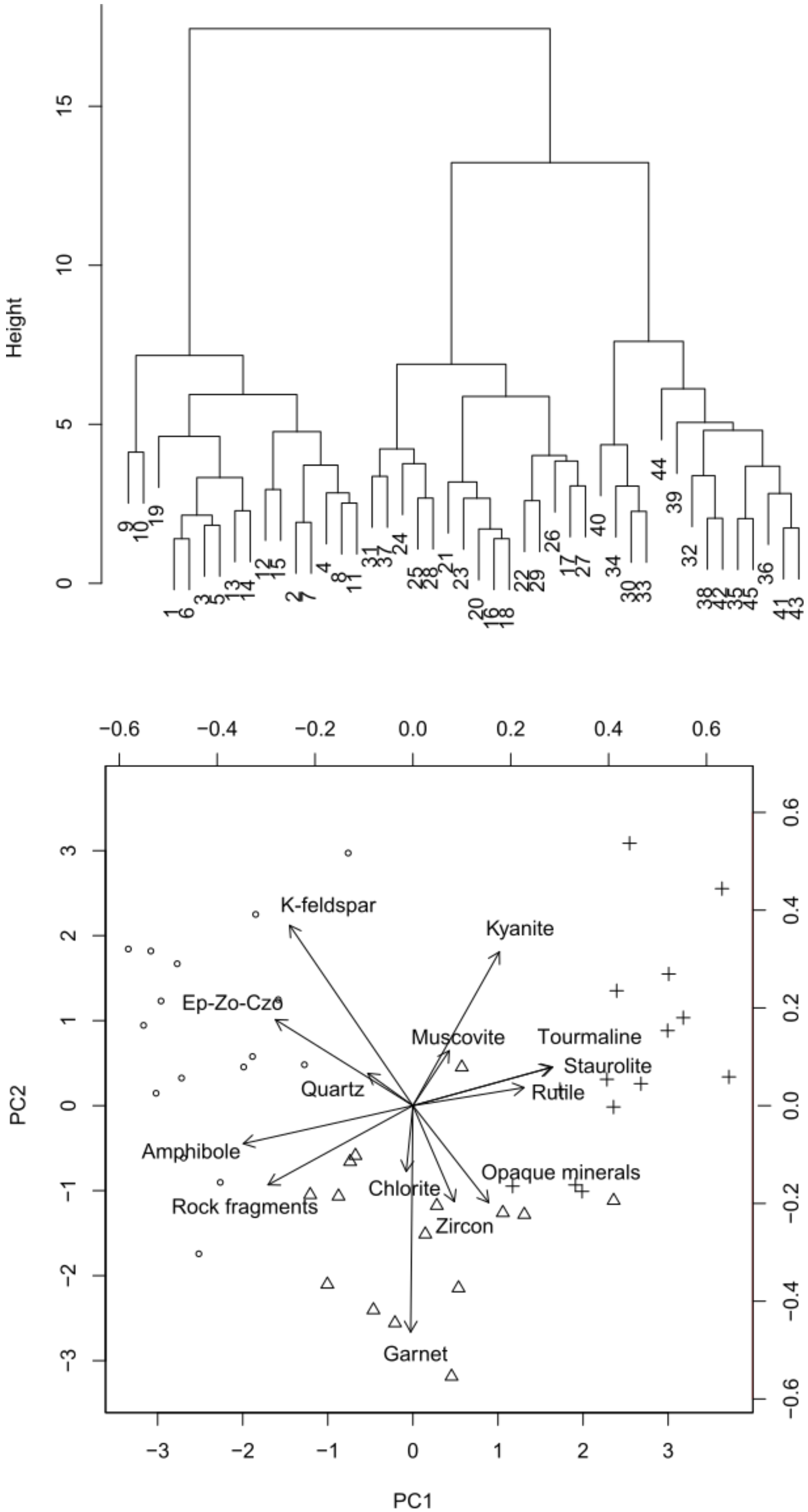

Figure 5: Cluster dendogram for samples (top) and compositional biplot with groups according to cluster analysis (bottom)

The cluster analysis of variables (**Figure 6**) yielded results similar to those of the PCA, yet with kyanite grouped with tourmaline and staurolite. This supports the assumption that the group marked with pluses represents the weathering process of metamorphic rocks. Garnet is grouped with chlorite and muscovite, which could all be products of the weathering of the Miocene clastites.

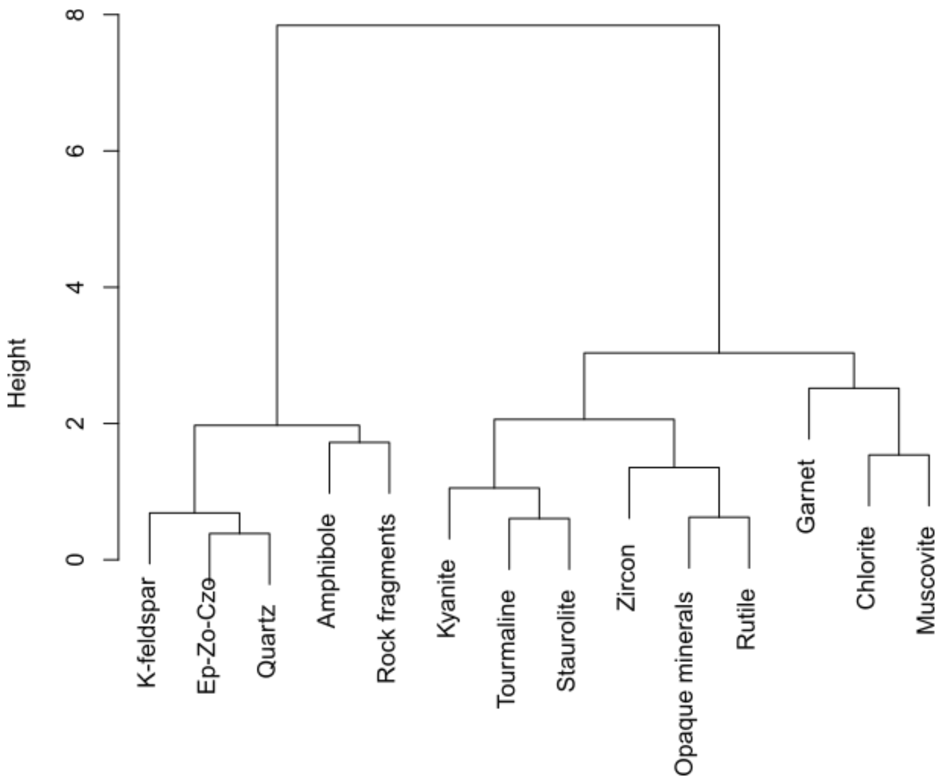

Figure 6: Cluster dendogram for variables

5. Conclusion

This study presents a detailed analysis of Quaternary sediments from the foothills of Medvednica Mountain, aimed at understanding their differences based on mineral associations and depositional environments. Through the use of compositional data analysis, including techniques such as cluster analysis and principal component analysis, the research has identified patterns in mineral distributions and associations, revealing significant geological complexities of the region. The findings indicate two main depositional environments: high-energy settings like rivers where heavier minerals are concentrated, and low-energy settings such as lakes and swamps where lighter minerals accumulate. These environments are reflective of a complex geological history that significantly influenced the observed mineralogical composition.

The study has confirmed findings of previos research that local rock weathering processes are the main driver in shaping the composition of Quaternary sediments in the region. This approach not only enhances our understanding of the geological framework of Medvednica Mountain but also demonstrates the application of compositional data analysis in geosciences, highlighting its value in addressing complex geological questions. However, there are limitations to this study, such as the limited number of samples and the constraints of the analytical methods used, which could affect the generalization of the findings. Future research should focus on expanding the sample size and incorporating advanced analytical techniques and dating to further refine our understanding of the depositional processes and mineral associations in this region.

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SAŽETAK

Kompozitna varijabilnost minerala na izdancima i u bušotinama neformalne jedinice formacije Bistra, Medvednica, Hrvatska

Ovo istraživanje bavi se varijabilnošću mineralnog sastava uzoraka neslužbene Bistra formacije na Medvednici u Hrvatskoj. Proučavani su kvartarni sedimenti iz izdanaka i bušotina. Koristeći modalnu analizu i statističke metode obrade kompozicijskih podataka (CoDa), cilj istraživanja bio je klasificirati uzorke i definirati povezanost varijabli kako bi se poboljšalo razumijevanje različitosti sedimenta. Analiza je otkrila značajne varijacije u sastavu minerala kao rezultat lokalnih uvjeta taloženja i trošenja specifičnih geoloških formacija. Ključni rezultati podrazumijevaju odredbu dvije različite sredine taloženja: visokoenergetska riječna, u kojoj se koncentriraju teži minerali, i niskoenergetska jezerska ili močvarna, u kojoj se akumuliraju lakši minerali. Korištenje analize glavnih komponenata i klaster analize dodatno je pojasnilo odnose među mineralima. Ovaj pristup pokazuje korisnost CoDa analize u geoznanostima, pružajući dublje uvide u podrijetlo i dinamiku taloženja kvartarnih sedimenata.

Ključne riječi: Bistra formacija; Medvednica; modalna analiza; obrada kompozicijskih podataka; klasifikacija

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Author's contribution

Anita Grizelj performed the fieldwork and provided the modal analysis. **Danijel Ivanišević** provided the statistical analysis and interpretation of the results.