

## GEOCHEMISTRY AND ORIGINE OF THERMO-MINERAL WATERS ON KOŽUF MOUNTAIN

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**A b s t r a c t:** The paper presents the results of geochemical surveys of the representation of trace elements in thermal mineral water from Kožuf Mountain and investigations relating to the representation of the stable isotopes of hydrogen and oxygen. The results displaying the representation of trace elements suggest that the water is enriched with arsenic, uranium and cesium, while other trace elements are within permissible concentrations. Studies on the representation of isotopes  $d^{18}O$ ,  $dD$  indicate that the values of stable isotopes are placed near the route of water from precipitation. This indicates that the water in primary reserve tanks is renewed with the water that comes from current rainfall.

**Key words:** geochemistry; origin thermal-mineral water; Kožuf Mountain

### INTRODUCTION

Thermal mineral waters within the volcanic complex appear in three locations: in the vicinity of the village of Mrežičko, in the locations of Topli Dol and Kisela Voda and in the locality “Alshar” in the location of Toplek. In these localities thermal mineral waters appear in the contact parts of the series of Triassic dolomites that in some places are intensively carsified and tectonically crushed, and Triassic and Cretaceous metamorphic rocks represented by different types of shale.

#### *Thermal mineral water Topli Dol*

There are multiple springs with different yield, spaced approximately 200 m on both sides of the river of Topli Dol. The main spring is located about 3 km southwest of the village of Mrežičko, on the road to Ržanovo, on the left side of the Topli Dol river. The spring is captured and it springs at the contact between shale and serpentine. The temperature of the water is 27°C, while its yield is 0.8 l/s (pH 6.5). In fact, there are three springs at a distance of 10 m at different levels in the river valley. The lowest is in the river bed itself which in this section is about 10 m wide. The middle spring has the largest spring yield and is 1 m

above the river, while the third one is 1.2 m higher than it. Iron deposits are found around springs, and as the water contains sulfur compounds it smells of sulfur. According to the content of macrocomponents (ionic composition), the water from the springs belongs to hydrocarbonate, calcium weakly magnesium waters.

#### *Thermal mineral water in the area of Mrežičko*

Approximately 1.5 km southwest of the village of Mrežičko on the left side of the road to Ržanovo there is a captured spring that springs from the slope diluvial deposits. The genesis of the water is vadose and is strictly of a local character, influenced by erosion processes. The eroded and mechanically shattered material that is precipitated here is permeable and it allows the infiltration of part of storm water. The result of this is a formed thermal mineral aquifer. The water obtains mineral properties by dissolving rocky masses through which ground water flows, while its sour taste comes from the content of  $CO_2$  absorbed by storm water. That increases the dissolving power of water which is another reason for its mineralization. The temperature of the water is 21°C with yield ranging from 0.07 to 0.1 l/s (pH 7.5).



The ring structures that are characteristic of this area complex also indicate this type of volcanism (Boev et al, 1990) (Fig. 2)

Geologically speaking the Kožuf area is built from several geological formations represented in several stratigraphic complexes as follows (Fig. 1):

- complex of Precambrian metamorphic rocks,
- complex of Paleozoic metamorphic rocks,
- complex of Triassic-Jurassic sedimentary rocks,
- complex of Upper Cretaceous sedimentary rocks,
- complex of Upper Eocene sediments,
- complex of Pliocene sedimentary and pyroclastic rocks,
- complex of Quaternary sediments.

Geological construction also includes complexes of magmatic rocks that are presented with:

- complex of metamorphosed rhyolite and pyroclastic rocks,
- complex of serpentine ultra-basic rocks,
- complex of basic magmatic rocks,
- complex of volcanic rocks.

All listed upper stratigraphic complexes basically make the complexity of the geological structure of the Mt. Kožuf, where the dominant place is still held by the Tertiary volcanic activity (6.5 to 1.8 MA Boev, 1988) with which numerous mineralizations are related, as well as post-volcanic phenomena that are manifested with the occurrence of thermal mineral waters are related.

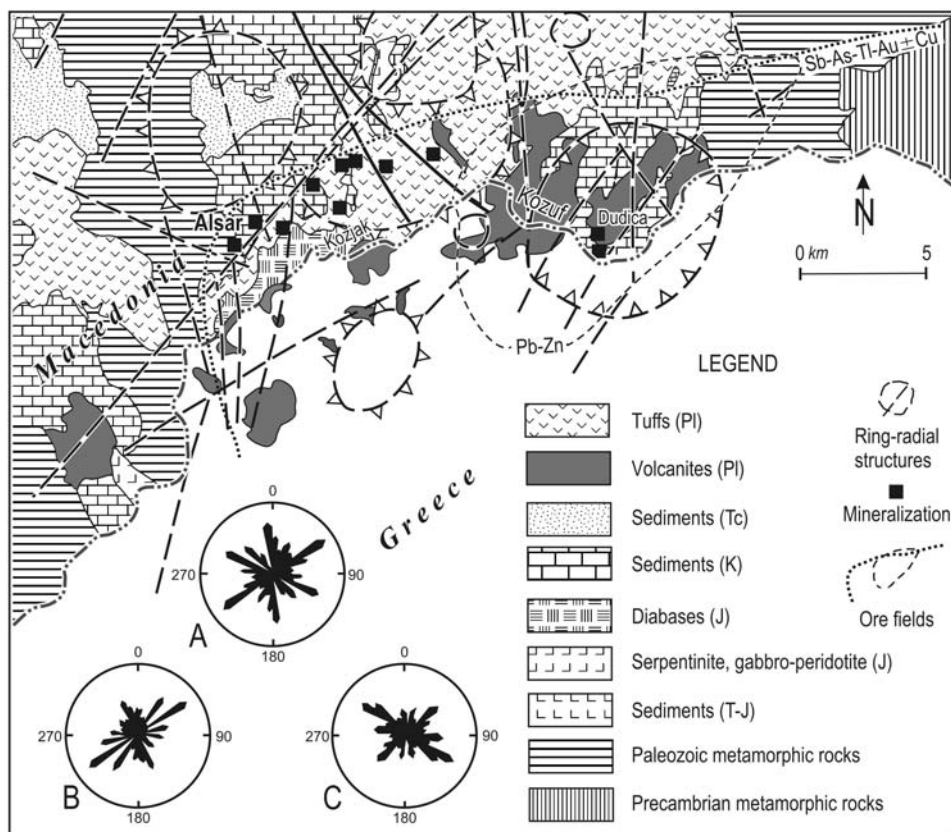


Fig. 2. The ring structures that are characteristic of this area (Boev, 1988)

## METHODOLOGY

For determination of trace elements in 15 samples collected from springs of thermal mineral water, 5 samples from each source (Topli Dol, Toplek, Kisela Voda) in 50 ml Nalgene plastic bottles and preserved with pure nitric acid to pH < 2. To determine the stable isotopes of hydrogen and

oxygen ( $d^{18}O$ ,  $dD$ ) 15 samples in 50 ml Nalgene plastic bottles were collected too.

The determination of the presence of trace elements was done by ICP-MS method and the determination of the presence of stable isotopes was done by ISO method.

## RESULTS AND COMMENTS

The results obtained from geo-chemical tests of the representation of macroelements (Mg, K, N, Si, Ca, Al) (Table 1) indicate that these are hydro-carbon, calcium poorly magnesium waters. It should be noted that the content of Na (Fig. 3), Mg (Fig. 4), Si (Fig. 5), K (Fig. 6), Ca (Fig. 7) is far greater in water samples from the vicinity of Mrežičko and Topli Dol compared to the water content in the vicinity of the village of Majdan. This is due to the fact that the thermal mineral water springs in the vicinity of the Majdan village has a much larger amount of water coming from the Majdanska River. This is due to the fact that these springs are in the very bed of the Majdanska River and consequently there is greater mixing of waters and as a result we have a reduced content of macroelements in the water from the thermal mineral water spring. The same conclusion can be derived when it comes to trace elements, in particular As (Fig. 8), Rb (Fig. 9), Sr (Fig. 10), Cs (Fig. 11), Ba (Fig. 12), U (Fig. 13). There is higher concentration of these trace elements in the water from

springs in the vicinity of the village of Mrežičko and of Topli Dol compared to the springs in the vicinity of the Majdan village.

The increased content of arsenic in the thermal mineral water from Kožuf Mountain in the amounts which are four times the allowable concentrations is due to the increased amount of arsenic in the rocks of Kožuf Mountain. It should also be mentioned that adjacent to these springs of thermal mineral water there is also a deposit site of arsenic and thallium from Alšar (Boev et al, 1993; Frantz et al, 1994; Ivanov, 1963; Kocnjeva et al, 2006; Kolios et al, 1980; Lepitkova, 1995; Volkov et al, 2006).

It is interesting to mention, of course, that the thermal mineral waters on Kožuf Mountain also contain increased concentrations of Cs and U. Increased concentrations of Cs and U are the result of the fact that volcanic rocks of Kožuf Mountain contain elevated concentrations of these elements (Boev, 1988).

Table 1

*Geochemical analysis of the thermal waters from the Kožuf area (ICP-MS method) ( $\mu\text{g/l}$ )*

Analyte symbol	Detection limit	K/1	R/1	T/1	MR/1	TD/1	MA/1	K/2	R/2	T/2	K/3	R/3	T/3	K/4	R/4	T/4
Na	5	35800	47900	2970	4030	5560	3050	38500	47300	2940	37200	47000	2890	38800	46400	4960
Li	1	210	270	<10	<10	4	<10	210	270	<10	205	270	<10	210	252	<2
Be	0,1	<1	<1	<1	<1	<0.1	<1	<1	<1	<1	<1	<1	<1	<0.5	0,6	<0.2
Mg	2	46300	39100	4660	6130	88900	5940	46400	39000	4830	46300	39000	4800	47200	36600	5070
Al	2	<20	<20	40	67	62	30	<20	<20	36	<20	<20	39	<10	<4	50
Si	200	31600	40500	6400	19700	12100	8200	35100	41200	6400	33900	41500	7000	35100	40100	7300
K	30	18100	15700	2860	3320	3220	1890	18400	12100	1170	39200	12300	1140	9440	11200	1130
Ca	700	215000	265000	20900	9600	22200	19600	218000	269000	21000	224000	269000	20800	238000	252000	21100
Sc	1	10	12	<10	<10	3	<10	11	12	<10	<10	12	<10	10	11	2
Ti	0,1	7,2	9,3	2,9	6,1	4,5	5,1	8,2	9	2,5	7,7	9,4	2,9	7,5	7,9	3,1
V	0,1	<1	<1	<1	2	0,8	<1	<1	<1	<1	<1	<1	<1	<0.5	<0.2	0,3
Cr	0,5	<5	<5	<5	<5	2,9	<5	<5	<5	<5	<5	<5	<5	<3	<1	<1
Mn	0,1	<1	85,2	1,7	1,6	4,8	1,3	4,6	86,6	<1	5,1	86,1	<1	4,7	83,2	1,4
Fe	10	<100	120	<100	<100	50	<100	<100	140	<100	<100	130	<100	<50	90	40
Co	0,005	<0.05	0,064	0,055	<0.05	0,09	<0.05	<0.05	0,094	<0.05	<0.05	0,071	<0.05	<0.03	0,05	0,036
Ni	0,3	<3	<3	<3	<3	5,7	<3	<3	<3	<3	<3	<3	<3	<2	0,9	<0.6
Cu	0,2	3,5	<2	3,7	10,1	1,1	7,6	7,4	2,1	4,8	2,8	<2	<2	1,6	0,4	0,8
Zn	0,5	23,5	5	11,9	<5	0,9	<5	11	<5	<5	13,6	<5	<5	3,1	1,2	<1
Ga	0,01	<0.1	<0.1	<0.1	<0.1	0,02	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.05	<0.02	0,02

Analyte symbol	Detection limit	K/1	R/1	T/1	MR/1	TD/1	MA/1	K/2	R/2	T/2	K/3	R/3	T/3	K/4	R/4	T/4
Ge	0,01	1,6	1,9	<0.1	<0.1	0,02	<0.1	1,9	1,8	<0.1	1,8	1,8	<0.1	1,85	1,76	<0.02
As	0,03	207	251	5,43	1,13	1,67	64	216	251	4,61	210	257	4,87	222	286	5,48
Se	0,2	<2	<2	<2	<2	0,6	<2	<2	<2	<2	<2	<2	<2	<1	0,6	<0.4
Rb	0,005	48,9	63,4	0,83	4,98	6,71	3,15	50,7	62,8	0,83	50,5	63,5	0,79	51,2	62,2	0,826
Sr	0,04	742	917	62,1	123	151	108	786	892	64,8	765	905	62,5	808	914	64,3
Y	0,003	0,38	0,33	0,17	0,15	0,089	0,07	0,3	0,33	0,11	0,3	0,33	0,1	0,31	0,318	0,136
Zr	0,01	0,1	0,2	0,1	0,4	0,59	0,1	0,1	0,1	0,1	0,2	0,1	0,1	0,15	0,06	0,12
Nb	0,005	0,1	0,27	<0.05	<0.05	0,051	<0.05	0,08	0,12	<0.05	0,115	0,14	<0.05	<0.03	<0.01	<0.01
Mo	0,1	<1	<1	<1	<1	3,5	<1	<1	<1	<1	<1	<1	<1	<0.5	0,2	<0.2
Ag	0,2	<2	<2	<2	<2	<0.2	<2	<2	<2	<2	<2	<2	<2	<1	<0.4	<0.4
Cd	0,01	<0.1	<0.1	<0.1	<0.1	0,02	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.05	<0.02	<0.02
In	0,001	<0.01	<0.01	<0.01	<0.01	<0.001	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.005	<0.002	<0.002
Sn	0,1	<1	<1	<1	<1	<0.1	<1	<1	<1	<1	<1	<1	<1	<0.5	<0.2	<0.2
Sb	0,01	<0.1	<0.1	<0.1	<0.1	0,25	10,6	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0,09	0,11	0,13
Te	0,1	<1	<1	<1	<1	<0.1	<1	<1	<1	<1	<1	<1	<1	<0.5	<0.2	<0.2
Cs	0,001	89,5	105	0,17	0,11	0,444	0,62	91,6	105	0,19	91,2	108	0,18	94	106	0,19
Ba	0,1	120	161	9,8	29,9	22	17,2	130	159	9,6	130	162	9,5	132	158	9,9
La	0,001	5,34	4,66	2,46	2,04	0,088	1,07	1,76	1,06	1,22	0,495	0,95	0,45	0,08	0,05	0,144
Ce	0,001	0,07	0,05	0,35	0,22	0,124	0,08	0,04	0,04	0,18	0,03	0,04	0,19	0,02	0,032	0,298
Pr	0,001	0,02	<0.01	0,05	0,06	0,027	0,01	<0.01	<0.01	0,03	<0.01	<0.01	0,03	0,005	0,006	0,036
Nd	0,001	0,06	0,03	0,17	0,22	0,115	0,07	0,05	0,02	0,12	0,045	0,04	0,11	0,045	0,026	0,154
Sm	0,001	0,02	0,01	0,04	0,06	0,022	0,01	0,01	0,01	0,02	<0.01	<0.01	0,02	0,01	0,01	0,028
Eu	0,001	<0.01	<0.01	<0.01	<0.01	<0.001	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.005	<0.002	<0.002
Gd	0,001	<0.01	<0.01	<0.01	<0.01	0,02	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.005	0,02	0,04
Tb	0,001	<0.01	<0.01	<0.01	<0.01	0,002	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.005	0,002	0,004
Dy	0,001	<0.01	<0.01	<0.01	<0.01	0,01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.005	0,02	0,02
Ho	0,001	<0.01	<0.01	<0.01	<0.01	<0.001	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.005	<0.002	<0.002
Er	0,001	<0.01	<0.01	<0.01	<0.01	0,01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.005	0,02	0,02
Tm	0,001	<0.01	<0.01	<0.01	<0.01	<0.001	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.005	<0.002	<0.002
Yb	0,001	0,02	0,02	0,02	0,02	0,01	<0.01	0,02	0,02	0,01	0,025	0,01	<0.01	0,015	0,018	0,014
Lu	0,001	<0.01	<0.01	<0.01	<0.01	0,002	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.005	0,002	0,002
Hf	0,001	<0.01	<0.01	<0.01	0,01	0,013	<0.01	<0.01	0,01	0,01	<0.01	<0.01	<0.01	<0.005	<0.002	0,004
Ta	0,001	<0.01	<0.01	<0.01	<0.01	0,016	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.005	<0.002	<0.002
W	0,02	<0.2	1	<0.2	<0.2	0,1	<0.2	<0.2	1	<0.2	<0.2	1	<0.2	<0.1	1	<0.04
Hg	0,2	<2	<2	<2	<2	<0.2	<2	<2	<2	<2	<2	<2	<2	<1	<0.4	<0.4
Tl	0,001	0,09	0,285	0,049	<0.01	0,005	0,401	0,103	0,268	0,043	0,103	0,271	0,026	0,104	0,256	0,062
Pb	0,01	2	2	1	1	<0.01	1	1	<0.1	<0.1	<0.1	<0.1	<0.1	0,5	<0.02	0,2
Bi	0,3	<3	<3	<3	<3	<0.3	<3	<3	<3	<3	<3	<3	<3	<2	<0.6	<0.6
Th	0,001	<0.01	<0.01	<0.01	0,025	0,06	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.005	<0.002	0,023
U	0,001	0,674	0,703	0,226	0,21	0,138	0,269	0,64	0,674	0,205	0,64	0,664	0,189	0,658	0,652	0,215

K/1, K/2, K/3, K/4 – Water samples from around Mrežičko; R/1, R/2, R/3, R/4 – Water samples from around Topli Dol; T/1, T/2, T/3, T/4 – Water samples from around Majdan; TD1 – Water samples from Topli Dol River; MR/1 – Water samples from Mrežička River; MA/1 – Water samples from Majdanska River;

The results of the tests on the presence of isotopes  $d^{18}O$  and  $dD$  are shown in Table 2 and Fig. 14 show the position of the calculated values of isotopes  $d^{18}O$  and  $dD$  in terms of the global direction of rain water (IAEA,1981, IAEA, 1983, IAEA, 1995). The position of the analyzed thermal mineral waters from Kožuf Mountain is near the direction of rain water which speaks of the fact that there is great renewal of thermal mineral waters with waters from current rain fall.

Table 2

Composition of the stable isotopes  $\delta^{18}O$  and  $\delta^2H$  in the thermal waters from the Kožuf area

Analyte symbol	$d^{18}O$	$dD$
Unit symbol	VSMOW	VSMOW
Analysis method	ISO	ISO
K/1-1	-9.64	-62.6
R/1-1	-9.7	-62.4
T/1-1	-9.53	-60.9
MR/1-1	-9.32	-59.5
TD/1-1	-8.38	-53.5
MA/1-1	-9.46	-58.6
K/2-2	-9.85	-62.6
R/2-2	-9.76	-62.4
T/2-2	-9.54	-59.9
K/3-3	-9.74	-62.8
R/3-3	-9.78	-62.5
T/3-3	-9.54	-60.7
K/4-4	-9.82	-63.2
R/4-4	-9.75	-62.6
T/4-4	-9.52	-60.7

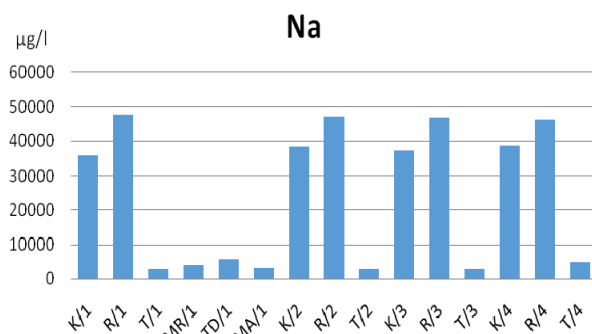


Fig. 3. Distribution of Na in mineral waters from Kožuf Mountain

K/1, K/2, K/3, K/4 – water samples from around Mrežičko;  
MR/1 – water samples from Mrežička River;  
R/1, R/2, R/3, R/4 – water samples from around Topli Dol;  
MA/1 – water samples from Majdanska River  
T/1, T/2, T/3, T/4 – water samples from around Majdan

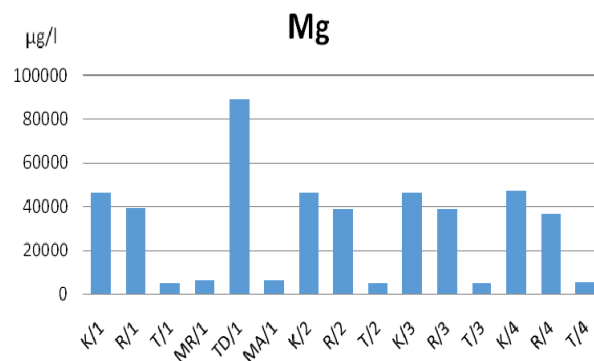


Fig. 4. Distribution of Mg in mineral waters from Kožuf Mountain

K/1, K/2, K/3, K/4 – water samples from around Mrežičko;  
MR/1 – water samples from Mrežička River;  
R/1, R/2, R/3, R/4 – water samples from around Topli Dol;  
MA/1 – water samples from Majdanska River;  
T/1, T/2, T/3, T/4 – water samples from around Majdan

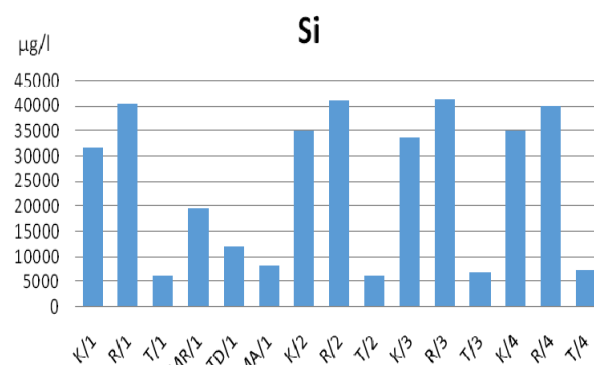


Fig. 5. Distribution of Si in mineral waters from Kožuf Mountain

K/1, K/2, K/3, K/4 – water samples from around Mrežičko;  
MR/1 – water samples from Mrežička River;  
R/1, R/2, R/3, R/4 – water samples from around Topli Dol;  
MA/1 – water samples from Majdanska River  
T/1, T/2, T/3, T/4 – Water samples from around Majdan

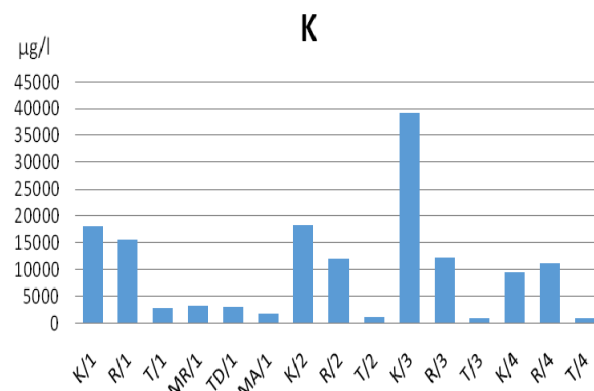
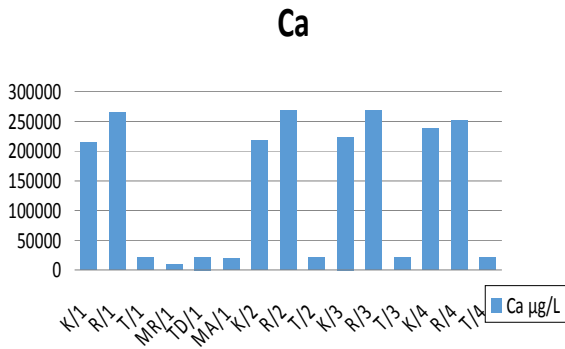


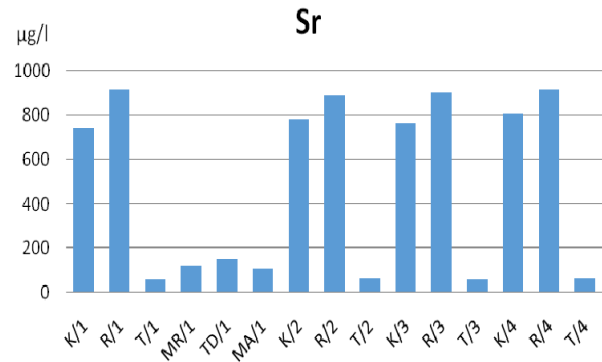
Fig. 6. Distribution of K in mineral waters from Kožuf Mountain

K/1, K/2, K/3, K/4 – water samples from around Mrežičko;  
MR/1 – water samples from Mrežička River;  
R/1, R/2, R/3, R/4 – water samples from around Topli Dol;  
MA/1 – water samples from Majdanska River  
T/1, T/2, T/3, T/4 – water samples from around Majdan



**Fig. 7.** Distribution of Ca in mineral waters from Kožuf Mountain

K/1, K/2, K/3, K/4 – water samples from around Mrežičko;  
 MR/1 – water samples from Mrežička River;  
 R/1, R/2, R/3, R/4 – water samples from around Topli Dol;  
 MA/1 – water samples from Majdanska River;  
 T/1, T/2, T/3, T/4 – water samples from around Majdan



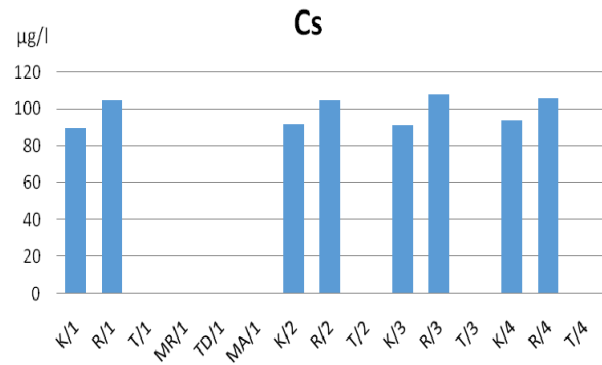
**Fig. 10.** Distribution of Sr in mineral waters from Kožuf Mountain

K/1, K/2, K/3, K/4 – water samples from around Mrežičko;  
 MR/1 – water samples from Mrežička River;  
 R/1, R/2, R/3, R/4 – water samples from around Topli Dol;  
 MA/1 – water samples from Majdanska River;  
 T/1, T/2, T/3, T/4 – water samples from around Majdan



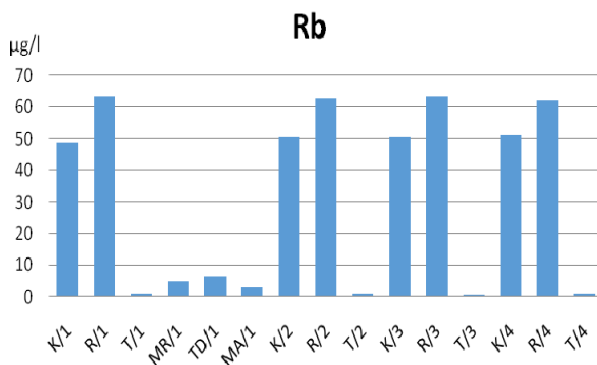
**Fig. 8.** Distribution of As in mineral waters from Kožuf Mountain

K/1, K/2, K/3, K/4 – water samples from around Mrežičko;  
 MR/1 – water samples from Mrežička River;  
 R/1, R/2, R/3, R/4 – water samples from around Topli Dol;  
 MA/1 – water samples from Majdanska River;  
 T/1, T/2, T/3, T/4 – water samples from around Majdan



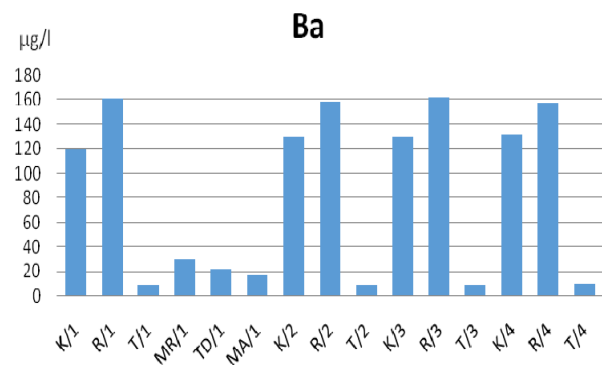
**Fig. 11.** Distribution of Cs in mineral waters from Kožuf Mountain

K/1, K/2, K/3, K/4 – water samples from around Mrežičko;  
 MR/1 – water samples from Mrežička River;  
 R/1, R/2, R/3, R/4 – water samples from around Topli Dol;  
 MA/1 – water samples from Majdanska River;  
 T/1, T/2, T/3, T/4 – water samples from around Majdan



**Fig. 9.** Distribution of Rb in mineral waters from Kožuf Mountain

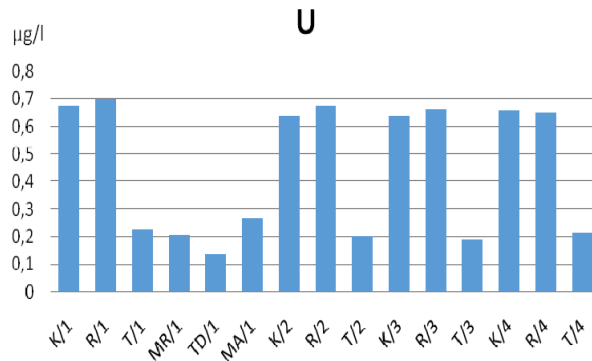
K/1, K/2, K/3, K/4 – water samples from around Mrežičko;  
 MR/1 – water samples from Mrežička River;  
 R/1, R/2, R/3, R/4 – water samples from around Topli Dol;  
 MA/1 – water samples from Majdanska River;  
 T/1, T/2, T/3, T/4 – water samples from around Majdan



**Fig. 12.** Distribution of Ba in mineral waters from Kožuf Mountain

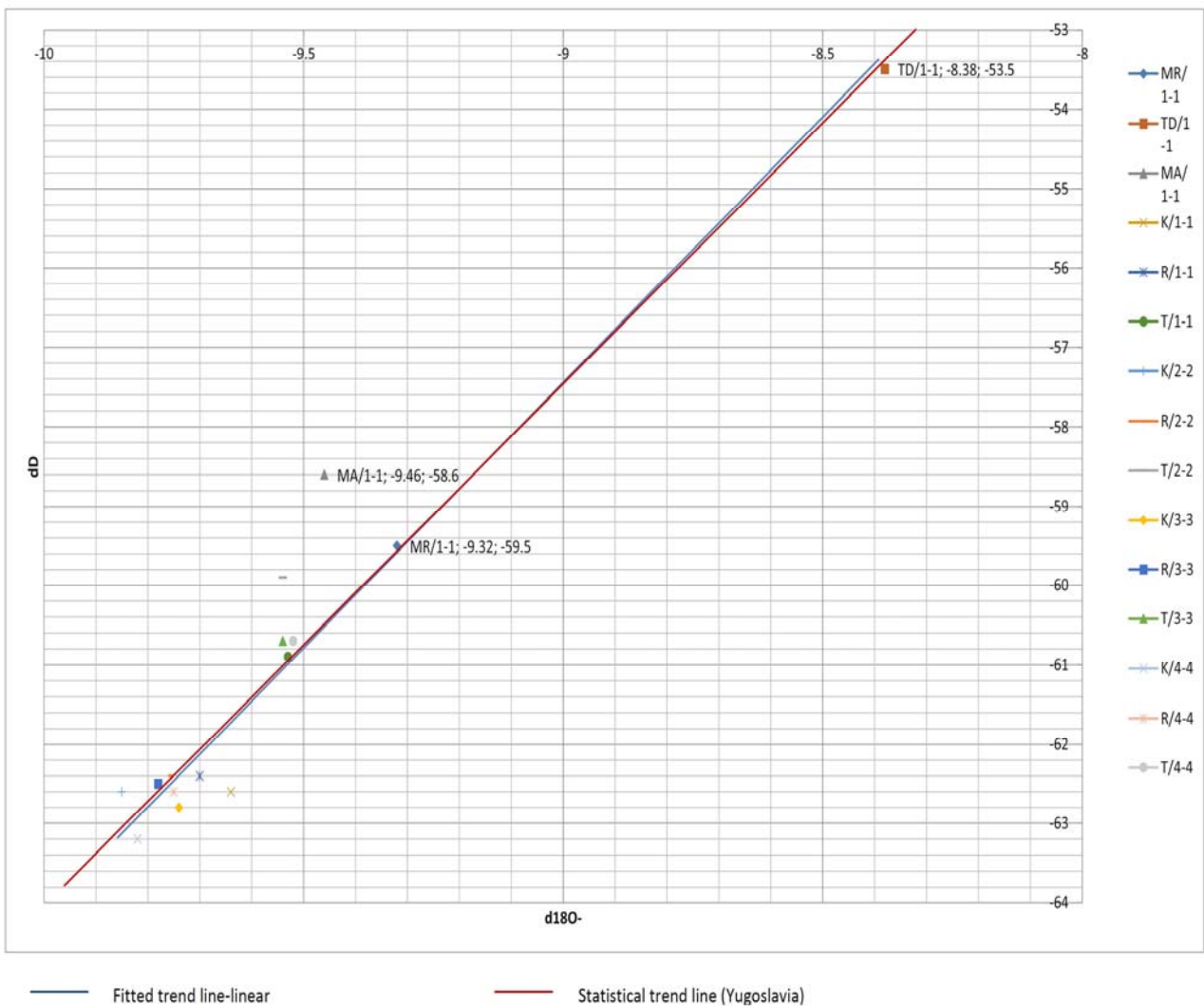
K/1, K/2, K/3, K/4 – water samples from around Mrežičko;  
 MR/1 – water samples from Mrežička River;  
 R/1, R/2, R/3, R/4 – water samples from around Topli Dol;  
 MA/1 – water samples from Majdanska River;  
 T/1, T/2, T/3, T/4 – water samples from around Majdan





**Fig. 13.** Distribution of U in mineral waters from Kožuf Mountain

K/1, K/2, K/3, K/4 – water samples from around Mrežičko;  
 MR/1 – water samples from Mrežička River;  
 R/1, R/2, R/3, R/4 – water samples from around Topli Dol;  
 MA/1 – water samples from Majdanska River;  
 T/1, T/2, T/3, T/4 – water samples from around Majdan



**Fig. 14.** Relation between  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  in the thermal waters from the Kožuf area and local direction of the current rainfall



## CONCLUSION

Geochemical investigations of the presence of macroelements and trace elements in thermal mineral water from Kožuf Mountain point to the fact that these are hydrocarbonic, calcium and poorly magnesium waters that have elevated concentrations of arsenic, cesium and uranium as a conse-

quence of the interaction of these waters with volcanic rocks. The results obtained for the presence of isotopes of  $d^{18}O$  and  $dD$  suggest that thermal mineral waters on Kožuf Mountain are mostly renewed with the water that comes from current rainfall.

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## Резиме

## ГЕОХЕМИЈАТА И ПОТЕКЛОТО НА ТЕРМОМИНЕРАЛНИТЕ ВОДИ НА КОЖУФ ПЛАНИНА

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**Клучни зборови;** потекло; термоминерални води; геохемија

Геохемиските испитувања на застапеноста на макроелементите и елементите во траги во термоминералните

води од Кожуф Планина укажуваат на фактот дека станува збор за хидрокарбонатни, калциумско-слабомагнези-

умски води во кои имаме зголемени концентрации на арсен, цезиум и уран како последица на интеракцијата на овие води со вулканските карпи. Добиените резултати за застапеноста на изотопите на  $d^{18}O$  и  $dD$  укажуваат на фак-

тот дека термоминералните води на Кожуф Планина во најголема мера се обновуваат со водите од современите врнежи.