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GEOCHEMISTRY AND ORIGIN OF PARTICLES PM-10 IN THE AREA OF TIKVEŠ, REPUBLIC OF MACEDONIA

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A b s t r a c t: We analyzed 13 aerosol samples collected on two sites in 2012: one in a commercial-industrial area and one in a residential area of Tikveš area. Coarse (PM 10) fractions were analyzed by ICP-MS at University in Štip. Seasonal gravimetric trends are obvious with higher concentrations in winter for both sides due to bad conditions of air circulation. Three main sources are proposed: (1) a natural source (crustal-geological formations) to which Mg, Li, Th, Na, Ca, U, Sr, Ca, Ti, V are related; (2) an industrial combustion source, which carries Cu, Mo, As, Zn, Pb; and (3) and nickel smelter source, characterized by high levels of Ni, Cr, Fe, Co, Mn.

Key words: PM-10 particles; trace elements; Tikveš; geochemistry.

INTRODUCTION

Among the valleys of Macedonia, which differ according to their geographical position, the Tikveš valley is characterized by specific geomorphological and anthropological-geographical features. With its surface of 2120 km^2, it covers a significant part of Macedonia. Its borders represent the Mariovo – Magelen Mountains on the south with range length of 1700 metres, on the west is the Mountain Borila with 1500 metres of length, on the south the Mountain Balija with 1400 metres length and Karadak with 750 metres height. Thus surrounded with mountains, Tikveš valley is intersected with the river of Vardar on the north, the river of Crna Reka on the west, and with the river of Luda Mara passing through its middle part (Fig. 1)

The geographical position and relief of Tikveš valley is a significant factor which affects the overall climatic characteristics. It is an area where two zonal climates have their effects: Continental and Mediterranean.

The Continental climate exists on the north and continues along the Vardar and Bregalnica rivers. As a result there are short but quite cold periods.

The Mediterranean climate affects the south coming from the Aegean Sea, bringing mild winters with relatively high temperatures.

The Tikveš valley is a rather warm area, which positively affects the development of winegrowing. The average temperature in Kavadarci is 18.9°C (with the highest temperature of 41°C), in Demir Kapija 19.5°C (with the highest temperature of 44.5°C). The warmest months in Kavadarci are July and August with average monthly temperature of 24.7°C, and the coldest month being January with average monthly temperature of 1.5°C.

The largest part of Tikveš valley is characterized by small amounts of precipitation. The area around Gradsko is considered to have the lowest precipitation in Republic of Macedonia. The average annual precipitation in Kavadarci is 484 mm.

The local inhabitants in Tikveš area, around 60 000, deal generally with agriculture (gardening, winegrowing and wine-production). About 100 million kilos of grapes are produced annually in this region.

A factory for nickel production was built in the Tikveš area in 1980. It produces 1,500,000 tons laterite types of nickel ore. By 2005, the complete amount of nickel ore originated from the Ržanovo mine, from the south parts of Tikveš valley, the Kožuf Mountain, and after 2005 ores from Albania, Turkey and Indonesia start to be reprocessed.

The work of this factory for nickel production affects the change of the mineral and geochemical structure of urban dust in the valley. The factory was built during 1976–1980, and it comprises equipment for reprocessing laterite ore from nickel with annual capacity of 2 million tons of ore. It annually produces about 16 000 tons of metal in the form of ferronickel, which contains Ni from 25 to 40 %. As a result a big amount of solid particles are generated, especially PM-10, which basically change the structure of urban dust (Boev et al., 2013; Stafilov et al., 2008, 2010; Bačeva, 2011; Stafilov et al., 2013).

The geological ingredients in the area of Tikveš involves various geological formations (Rakičević and Hristov, 1965) with different geological age (Fig. 1). The oldest formations have a NW-SE direction and belong to the inner part of the Vardar zone. The lowest Paleozoic (Pz) metamorphic layer consists of two series amphibolites and amphibole-chloritic shale with marble pro-layers and quartz-sericite shale with marble pro-layer and phyllites. In the structure of the Vardar zone there is presence of serpentinites.

Over the Paleozoic formations the Mesozoic formations developed (Mz), mainly in the lower chalk zone. The Turonian sandstones (K2), conglomerates and massive chalkstone stretch southwest and western part of the Tikveš area. The diabase and submarion outpouring of spilite are also common in the lower part of this sequence, where there are smaller masses of gabrous. The Paleozoic and Mesozoic rocks cover almost 40 km^2 from the west and south-west part of Tikveš area.

Uppereocenic (E_3) flysch sediments and yellow sandstones are present along the valleys of the Vardar, Crna Reka and Luda Mara rivers, as well as in a small amount in the Tikveš basin. These sediments with 3900 m depth cover 35 m^2 of the northern part of the area.

The Tikveš basin is filled with Pliocene (Pl) sediments represented with various series of sandstone. The Pliocene sediments cover the largest part of the area (about 190 km^2).

Southeast of Kavadarci there are Quaternary (Q) pyroclastic vulcanic rocks represented by tufs, brecia-sand conglomerates, which cover about 25% of the area. The Quaternary period is represented with diluvium (d), river terraces (t) and alluvium (al).

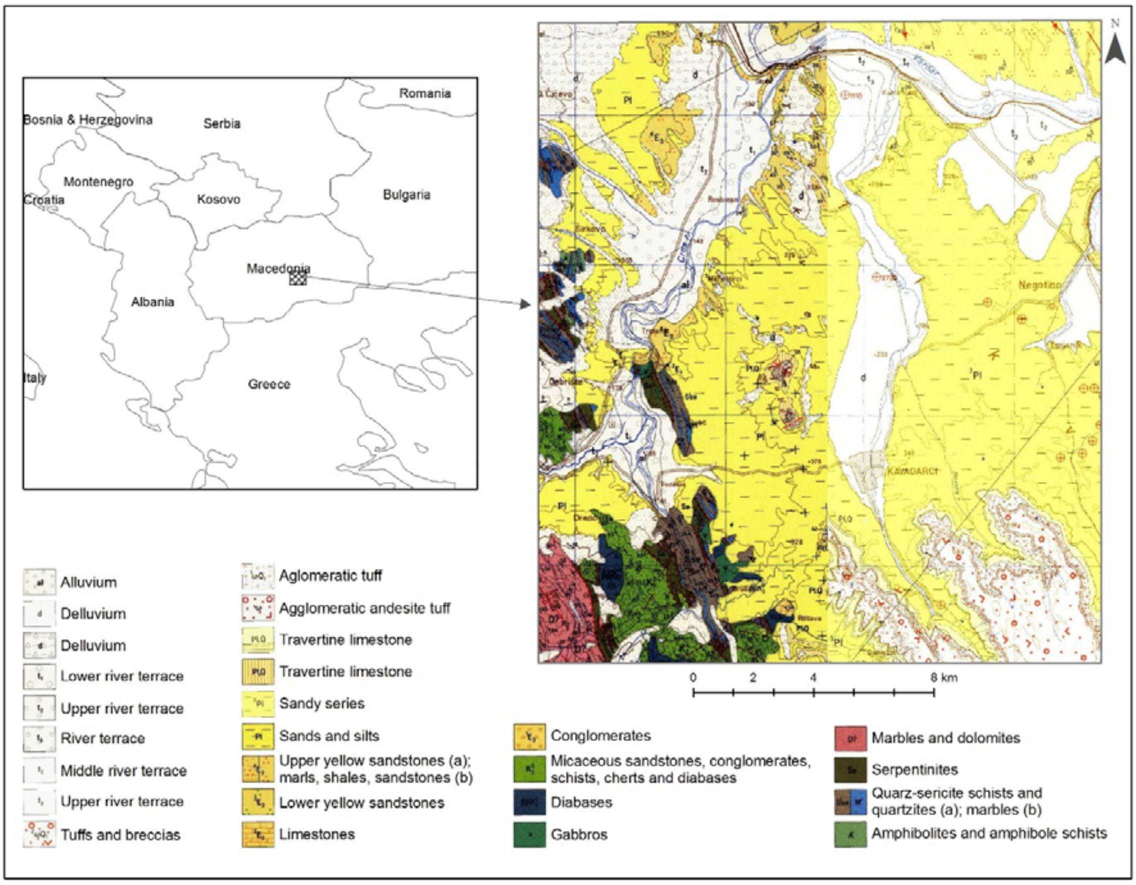

Fig. 1. Geographical position and geological map of Tikveš area

WORK METHODOLOGY

Different terms are used for the description of the origins and characteristics of particles. There is a tendency for random use of terminology, which has different meaning in popular and scientific context. Here are some of them:

Suspended solid particles (*SPM*) and *totally suspended particles* (*TSP*). Both terms denote the total number of particles in the air; they are often measured by taking samples with larger volume without selection of the quantity of the take-in air.

Solid particles (*PM*). Sometimes this term is used as a shortened form, but more often as PM_{10} or $PM_{2.5}$. PM_{10} is the mass concentration of particles (PM) which is due to the particles that pass through the selected air entrance, and have got 50% efficiency of particles with aerodynamic diameter of 10 μm. $PM_{2.5}$ is a concentration of particles with diameter of 2.5 μm.

Fine particles. These are particles with diameter smaller than a few μm. Sometimes the synonym $PM_{2.5}$ is used.

Ultrafine or nanoparticles are those that are smaller than 0.2 μm, whose size is expressed in nm (nanometres).

Aerosols are all solid and liquid particles suspended in the air.

Large pieces of dust. They are pieces of material which are formed during a mechanical process such as grinding or crushing. The size can sometimes be defined.

Dust. Smaller particles than the previous ones. Usually bigger than 1 μm.

Smoke. Particles formed during incomplete combustion as a combination of carbon and condensed volatile materials. They are usually smaller than 1 μm.

Dark smoke. Suspended particles which are determined by the reflexive dyeing method. The size is not determined, but latest measurements have not showed particles bigger than 10 μm aerodynamic diameter, and 50% particles with diameter of about 4 μm. These measurements may correspond to respirable fraction.

ACGIH and ISO conventions. Human respiratory system during evolution has enabled itself to filtrate the bigger particles in the early phase, and the percentage of particles that reach the lungs largely depends on the size of the particle. The American Conference of Governmental Industrial Hygienists (ACGIH) and the International Organization for Standardization have defined the fraction of particles according this ability of human respiratory system.

Gathering samples is done according to standard procedures by setting up two mobile stations, one in the area of the village of Vozarci (near the iron smeltery for ferronickel) and the other in the urban part of the town of Kavadarci. 10 samples have been collected in the area of the village of Vazarci, and 13 from the urban part of Kavadarci.

The determination of concentration of elements in traces is performed applying the ICP-MS method with accordance to ISO standards.

RESULTS AND DISCUSSION

The results from the analyses of the presence of elements in traces in particles PM-10 from the Tikveš area, are presented in Table 1 (urban part of the town of Kavadarci) and Table 2 (surrounding area of the village of Vozarci in the vicinity of the ferronickel smeltery).

The results regarding the quantitative presence of particles PM-10 in the urban part of the town of Kavadarci and surrounding area of the village of Vozarci are presented in Table 3. The results point out that the quantitative presence of particles PM-10 is far greater in the surrounding area of village of Vozarci (Fig. 2) (surroundings of ferronickel smeltery) compared to the urban part of the town of Kavadarci (Bačeva et al., 2011).

The distribution of the presence of particles PM-10 in the surrounding area of village of Vozarci in a time span of 24 hours is shown in Fig. 2. From the diagram we can deduce that the emission of hard particles PM-10 during 24 hours cycle of measurements is very volatile, with periods when the concentration of particles PM-10 ranges from ~50 μg/m^3 till 800 μg/m^3.

The distribution of elements of traces in particles PM-10 in the Tikveš area is presented with the following diagrams (Figs. 3, 4, 5, 6, 7).

The diagram in Fig. 3 shows increased presence of nickel in the dust in almost all locations in the area of Vozarci, in contrast to presence of nickel in the area of Kavadrci, where its presence is significantly lower, with some variance in location (2). Likewise, higher presence of iron and aluminum is found in the region of Vozarci, compared to the region of Kavadarci. The presence of other microelements in Vozarci is also somewhat higher, excluding chrome, which is more present in the dust from the region of Kavadarci (Solomons (1995; Bačeva et al., 2011; Boev et al., 2013, Dudka et al., 1997).

Figure 4 diagram shows that from the macroelements the presence of sodium is the highest in the dust, both in Kavadarci and Vozarci, without significant variance in various locations, excluding location nr. 2 in Kavadarci, where the presence of sodium is lower, contrary to other microelements such as calcium and magnesium, the presence of which is significantly higher in the dust, compared to other locations in Kavadarci. The presence of these elements, which have purely lithogenic origin, is related to the geological composition of the terrain (Šajn, 1999; Šajn, 2000; Stafilov et al., 2013).

If location 2 is taken into account, the sum of average values of macroelements is higher in Kavadarci. If this place is excluded, the sum of average values of macroelements is almost identical to that of Vozarci. Regarding other elements, the presence of sodium is the highest in the dust, both in Kavadarci and Vozarci, followed by magnesium, calcium, and potassium with the lowest presence. (Fig. 5)

T a b l e 1

Elements of traces in particles PM-10 from the urban part of Kavadarci (mg/kg)

	1	2	3	5	6	7	8	9	10	12	13	14	15
Na	2044	857	687	2159	1496	1614	1932	800	1417	1608	1782	979	1773
Mg	109	3212	140	306	156	132	152	97	121	99	132	344	123
Ca	71	4563	72	252	41	37	67	24	14	8	76	269	54
K	21	25	39	78	52	23	26	114	65	1	37	115	22
Fe	2545	71783	28369	35424	37572	20757	18916	1136	7178	23901	22887	25345	6131
Al	679	2911	5747	10160	9263	4942	1777	687	893	2425	7009	13140	798
P	1244	60202	850	3963	137	94	523	1283	1474	915	764	5509	2434
Ti	230	15699	428	714	416	359	301	30	42	154	429	818	297
B	4781	6302	8675	5045	6135	3212	4544	3745	1225	6773	2589	6990	5087
V	<10	<10	<10	45	39	10	<10	<10	<10	<10	37	38	<10
Cr	760	804	898	1300	1135	958	1103	750	880	925	991	1440	1053
Mn	165	1144	163	489	418	126	40	382	379	89	249	503	56
Ni	415	917	1375	3501	1541	2269	691	288	1921	765	580	820	9571
Co	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Cu	627	2424	2855	971	2581	575	2326	2380	648	2473	944	457	307
Zn	7520	7487	8948	13698	8620	10939	13832	7944	18470	8708	16875	2868	5040
Ga	18	170	14	37	37	19	10	20	<10	16	20	46	22
As	<10	<10	<10	177	<10	<10	<10	<10	<10	<10	<10	120	<10
Cd	57	<5	<5	<5	<5	<5	<5	<5	<5	<5	6	<5	<5
Rb	<10	<10	12	33	27	<10	<10	<10	<10	<10	16	53	<10
Sr	57	48442	120	552	151	128	101	179	155	187	256	637	158
Ba	42	4145	<10	500	319	138	<10	286	<10	<10	197	921	162
Pb	345	<10	54	1446	88	4423	<10	<10	<10	25	290	376	18
Bi	74	662	85	108	69	55	52	-82	-82	53	83	81	65
Sn	599	551	420	493	1570	570	352	443	283	779	308	219	34
Li	64	272	39	138	65	<10	38	<10	<10	<10	136	37	87
Sb	<10	<10	<10	<10	<10	222	<10	<10	<10	<10	<10	13	<10
Ag	13	27	15	23	81	17	<10	<10	<10	<10	23	48	62
Th	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
U	<5	<5	<5	15	<5	6	9	<5	12	<5	18	15	6
Be	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
Ge	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Mo	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Pd	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Cs	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Tl	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5

T a b l e 2

Elements of traces in particles PM-10 from the surrounding area of the village of Vozarci (mg/kg)

	1	2	3	4	5	6	7	8	9	10
Na	1621	1333	1174	1006	1000	977	851	891	788	817
Mg	1009	725	830	615	577	563	470	439	396	377
K	160	32	77	18	67	107	14	37	36	4
Ca	340	206	312	169	133	139	113	126	89	71
Fe	72930	48153	55640	51978	67568	24636	14612	18226	12369	8744
Al	44755	29666	43311	25702	24181	20546	16611	16261	17857	13727
P	6437	2726	3033	1965	1988	717	426	1839	80	65
B	7747	13970	6636	4435	1155	3788	1412	228	6402	344
Ba	26391	24941	1061	567	15672	858	25529	558	6228	1216
Ti	1411	663	1453	800	481	446	244	280	193	1274
V	68	<10	28	<10	<10	<10	<10	<10	<10	<10
Cr	930	334	525	670	434	27	16	<10	<10	418
Mn	1740	283	911	310	703	174	242	8247	101	11
Co	63	27	44	13	63	27	14	11	<10	<10
Pb	340	2	326	149	2007	927	262	297	166	881
Bi	459	428	117	147	239	130	117	34	328	220
Ni	8525	30325	22697	6887	50474	40566	34280	7693	11510	6595
Cu	1643	2646	2760	1309	1774	1520	6409	2513	1456	1384
Zn	10480	10833	15121	5646	12148	10252	10224	9339	3291	4781
Ga	1269	1169	74	44	648	42	1085	40	270	62
As	<10	<10	141	13	<10	<10	18	31	34	51
Rb	98	41	46	19	34	64	10	17	22	4
Sr	612	451	745	476	339	495	359	495	246	224
Ag	254	218	67	29	86	55	54	49	55	120
Cd	<5	<5	<5	<5	78	144	<5	<5	<5	35
Sn	<10	<10	3699	<10	1352	<10	<10	<10	<10	<10
Sb	<10	68	<10	<10	<10	<10	<10	<10	<10	<10
Th	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
U	22	17	12	18	28	13	11	34	17	15
Li	234	117	90	12	188	614	244	168	63	14
Be	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
Ge	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Mo	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Pd	<10	<10	<10	<10	49	<10	<10	<10	<10	<10
Tl	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
Cs	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10

Table 3

Calculation of the presence of PM -10 in the analyzed area

Type of the filters = 37 mm PVC + Cell.SP, h = 24, min = 1440, Q = 2, Vl = 2880. V = 2,88 m^3

Locattion	Type of the sampler	Date 2012	Number of the filters	M_1	m_2	PM10 µg/m^3
Vozarci	37 mm Case Cassela	20 IX	1	0.3271	0.3284	451
		21 IX	2	0.325	0.3261	35
		22 IX	3	0.3192	0.3213	729
		23 IX	4	0	0	0
		24 IX	5	0.3352	0.3375	799
		25 IX	6	0.3222	0.3237	521
		26 IX	7	0.3332	0.3354	764
Kavadarci	37 mm Cyclon	20 IX	8	0.3262	0.3272	347
		21 IX	9	0.332	0.3322	69
		22 IX	10	0.331	0.3313	104
		23 IX	11	0	0	0
		24 IX	12	0.3321	0.3323	69
		25 IX	13	0.3411	0.3422	382
		26 IX	14	0.3376	0.3383	243
			15	empty (празен)		
Vozarci	37 mm Case Cassela	27 IX	1	0.338	0.3403	799
		28 IX	2	0.325	0.3284	1181
		29 IX	3	0.333	0.3346	556
		30 IX	4	0.327	0.3285	521
		01 10	5	0.3302	0.3319	590
Kavadarvi	37 mm Cyclon	27 IX	6	0.337	0.3372	69
		28 IX	7	0.328	0.3281	35
		29 IX	8	0.3211	0.3231	694
		30. IX	9	0.3272	0.329	278
		01 X	10	0.325	0.3251	35

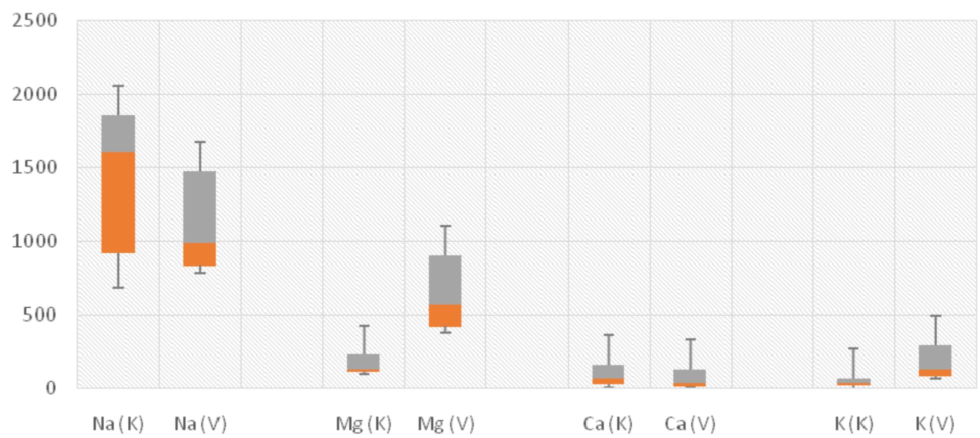

Fig. 2. Concentration of the PM-10 particles in the Tikvešh area
(K – Kavadarci, V – Vozarci)

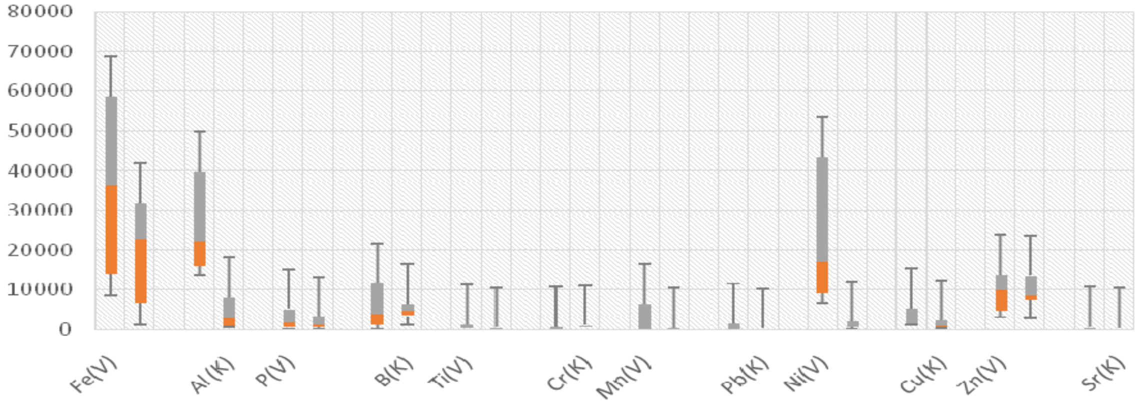

Fig. 3. Macro- and microelements in particles PM-10 from Vozarci and Kavadarci
(K – Kavadarci, V – Vozarci)

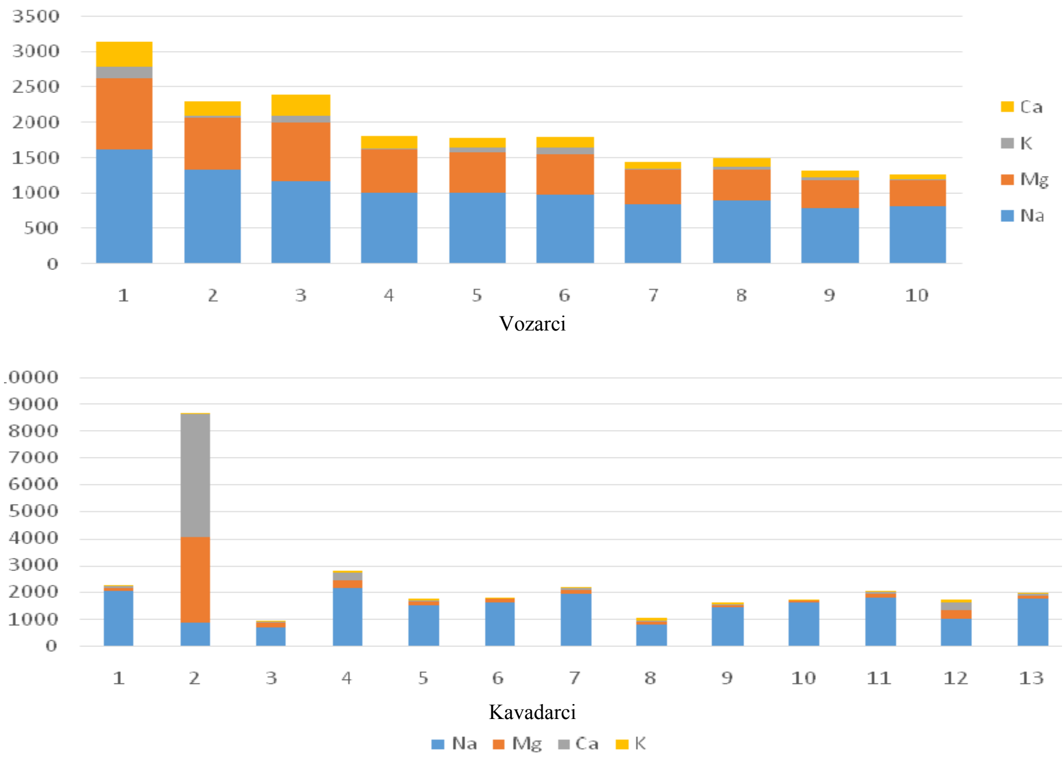

Fig. 4. Distribution of macroelements in particles PM-10 in the Tikveš area

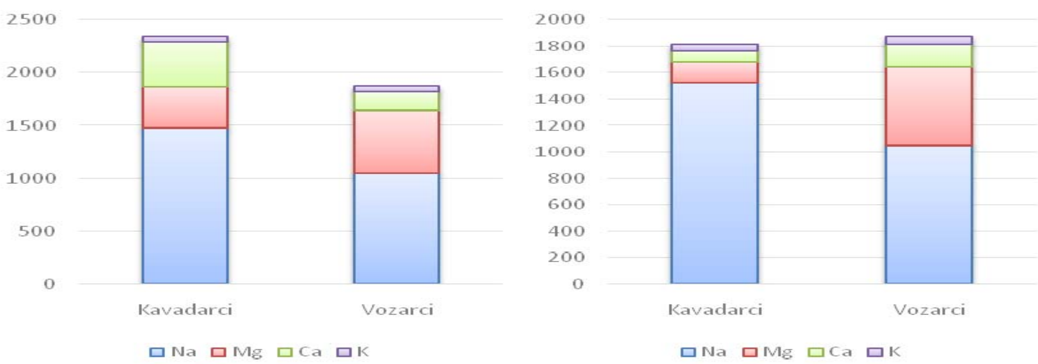

Fig. 5. Diagram of average values of macroelements in particles PM-10

The distribution of the sum of microelements in Kavadarci and Vozari is shown in the corresponding charts. Vozarci has a higher overall presence of microelements compared to Kavadarci, excluding one location (2) in Kavadarci, where the sum of microelements is almost identical to that of Vozarci (Fig. 6).

This chart also shows that Vozarci region has a far higher overall presence of microelements, compared to Kavadarci, including aluminum, iron, manganese, barium and nickel, but excluding chrome, which is more present in the dust of the region of Kavadarci (Fig. 7).

The analysis also points out that the presence of elements: Mg, Li, Th, Na, Ca, U Sr, Ti, V, which have lithogenic origin, is almost identical in the Tikveš area, and it can be concluded that the presence of these elements stems from the geological composition of the terrain.

The presence of elements Rb, K, Cs, Fe, P, Ba, Mn, Ni, Cr, Co, Zn, Sn, Pb, Cu, Mo, Cd, As, Ag, Sb has anthropogenic nature and it can be deduced that this presence is far greater in the surroundings of the village of Vozarci, in the vicinity of the ferronickel smeltery (nickel ore processing). (Boev et al., 2005; Maksimović, 1982).

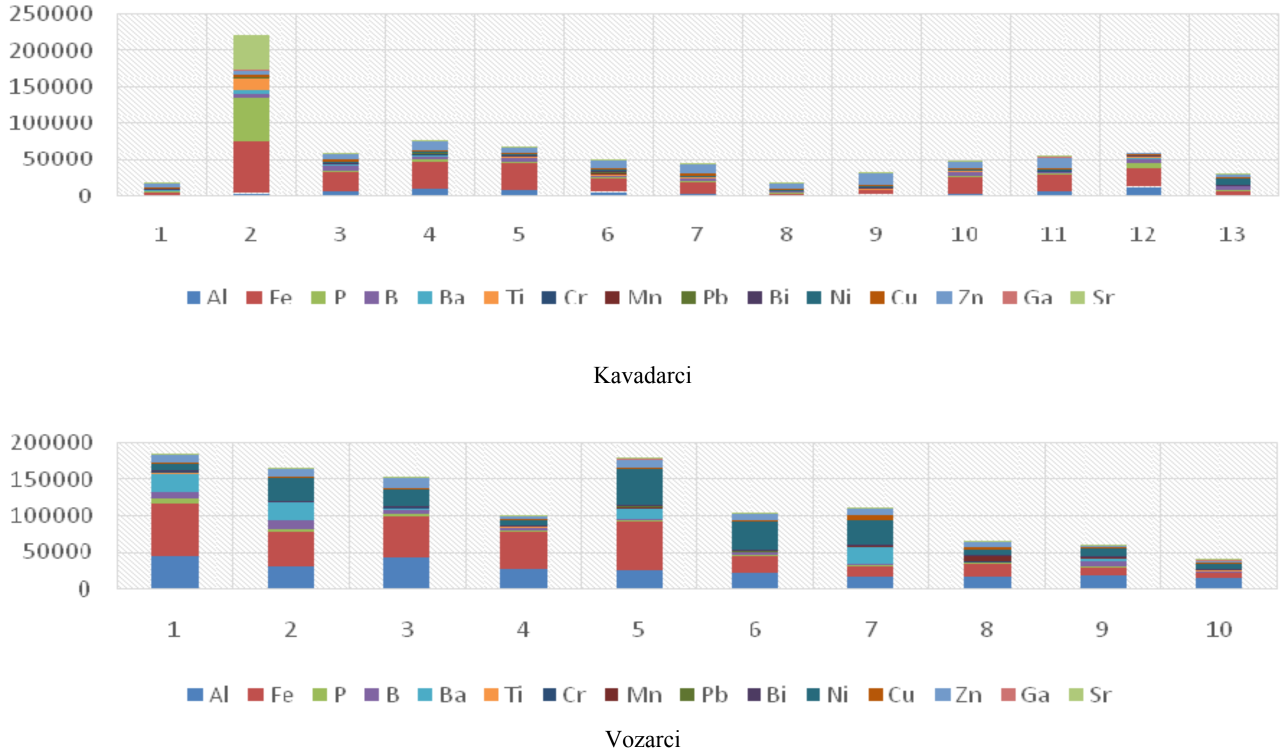

Fig. 6. Diagram of distribution of elements in the traces of particles PM -10

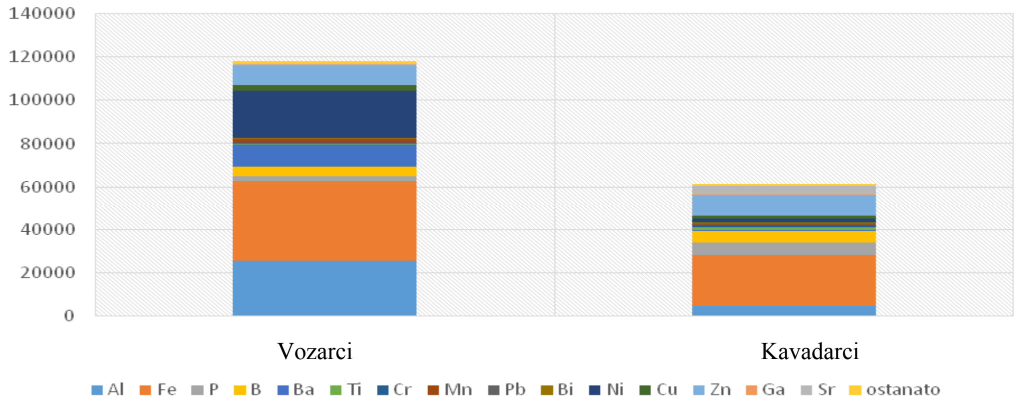

Fig. 7. Diagram of sum values of elements in traces

CONCLUSION

The results from this paper show that the surroundings of the village of Vozarci in the Tikveš area have significantly increased presence of particles PM-10. The analyses of the elements in the traces, point out to three geochemical groups of elements stemming from different sources:

– Elements form the group of Mg, Li, Th, Na, Ca, U, Sr, Ca, Ti, V have typical lithogenic origin, and their presence in particles PM-10 is related to the geological composition of the terrain.

– Elements from the group of Cu, Mo, As, Zn, Pb have typical anthropogenic origin and their presence in particles PM-10 can be related to the burning of fossil fuels and emission of exhaust gases from motor vehicles.

– Elements form the group of Ni, Cr, Fe, Co, Mn have typical anthropogenic origin and their presence is a direct consequence of the work of the ferronickel smeltery.

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ГЕОХЕМИЈА И ПОТЕКЛО НА ЧЕСТИЧКИТЕ ПМ-10 ВО ОБЛАСТА ТИКВЕШ, РЕПУБЛИКА МАКЕДОНИЈА

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Клучни зборови: честички ПМ-10; елементи во траги; Тиквеш; геохемија

Од прикажаните резултати во овој труд може да се констатира дека во областа Тиквеш во околината на селото Возарци има значително зголемена концентрација на честички ПМ-10. Испитувањата на елементите во траги укажуваат на три геохемиски групи на елементи кои имаат различно потекло, и тоа:

– Елементите од групата на Mg, Li, Th, Na, Ca, U, Sr, Ca, Ti, V имаат типично литогено потекло и нивната зас-

тапеност во честичките ПМ-10 се поврзува со геолошката градба на теренот,

– Елементите од групата на Cu, Mo, As, Zn, Pb имаат типично антропогено потекло и нивната застапеност во честичките ПМ-10 може да се поврзе со согорувањето на фосилните горива и со емисијата на издувните гасови од моторни возила.

– Елементите од групата на Ni, Cr, Fe, Co, Mn имаат типично антропогено потекло и нивната концентрација е директна последица на работата на топилницата на феро-никел.