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## ASSESSMENT OF THE WATER QUALITY OF THE RIVER VARDAR IN TERMS OF THE IRRIGATION IN SKOPJE VALLEY IN THE REPUBLIC OF MACEDONIA

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#### ABSTRACT

Skopje Valley is the second largest and also the most populous region in the country. Waste water from households, industry and agriculture ends up in the Vardar River as the largest drainage artery in the region in the republic. Due to the international conventions for the protection of international watercourses and therefore benefiting from irrigation water, the quality of Vardar River in Skopje and elsewhere in R. Macedonia is permanently active topic under constant surveillance. The waters of Vardar through the Skopje string section according to the legal regulations of the Republic Macedonia, shows solid summative quality. More worrisome is the situation with the organic loads and heavy metals that reduce the irrigation eligibility for agricultural purposes.

Keywords: Skopje Valley, Vardar, Measuring Point, Pollution, Heavy Metals, etc.

#### **INTRODUCTION**

Vardar River Basin covers 80% of the territory of the Republic Macedonia [13]. It is also the largest recipient of industrial waste waters, as well as the largest contributor of water for irrigation of all possible runoff in the country. Rivers are the vital source of water, which plays an important role in development of nation and sustenance of life [2]. Available water resources followed by numerous water management problems in the territory of the Skopje Valley and in the entire basin inevitably influenced the water quality in the recipient. An array of natural and anthropogenic factors have changed the water quality of Vardar River in Skopje Valley, which arise questions about its value and use for agricultural purposes. From these reasons, spontaneously appeared the need to meticulously study the water quality of Vardar River in Skopje Valley whose water can be used for irrigation of Skopsko Pole. In this paper were studied the waters of Vardar River in Skopje Valley which were monitored at two measuring points: at the entrance to the valley downstream from the village Radusha

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(point 1, fig. 1) and the area near to Jurumleri (point 2, fig. 1). The assessment of the water quality of Vardar was based on 11 parameters of the basic physical and chemical analysis and 8 heavy metals as a criterion for quality that have a significant impact on agricultural production, during the irrigation conditions. The purpose of the paper is to systematically detect the actual quality, contamination and spatial distribution of heavy metals in the water flow and to identify natural and anthropogenic sources of pollution in order to propose the most appropriate measures to prevent pollution and improving water quality.

#### STUDY AREA

Skopje Valley is situated in the northern part of the Republic Macedonia. It occupies the upper basin of the Vardar Riverand the territory bounded by mountains: Goleshnica, Karadjica, Suva Montain, Suva Gora, Zeden, Skopska Crna Gora and the Gradishtanska Mountain [3]. Skopje Valley covers the lower catchment areas of some of the major tributaries (Treska, Lepenec, Pchinja) and also the catchment areas of some of the smaller tributaries like (Serrava, Markova River). In this framework the Skopje Valley covers 1924.2 km<sup>2</sup> or 7.5 % of the territory of the Republic Macedonia and in the flat strip to 300 meters above sea level are accounted 343.9 km<sup>2</sup> (fig. 1).

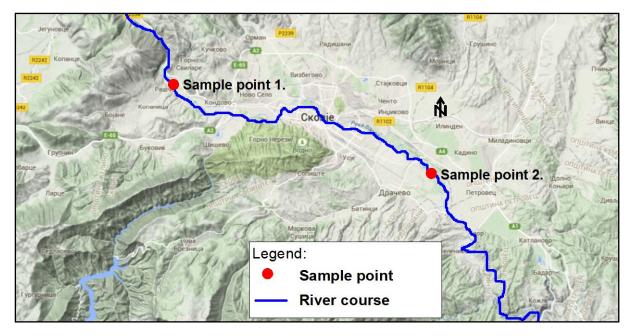


Figure 1. Vardar River through the Skopje Valley

#### **METHODS AND MATERIALS**

Regarding the need for more objective detection of the state of pollution the water in the Vardar River in Skopje Valley, during the season 2009/2010 (from September, 2009 to August, 2010), were purposefully selected two points of the water quality assessment whose monitoring was conducted at a frequency of analyzed samples once a month. The choice of

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the measurement points was dedicated executively, starting from the practical agricultural purposes. The first, upper monitoring point downstream from the village Radusha was determined in order to assess which input in Skopje Valley is entered from the upstream of the Polog region, and the lower monitoring point near the village Jurumleri is chosen in order to assess what changes undergone in the waters of Vardar in the Skopje region and its suitability for irrigation. Sampling was carried out at the measurement point nearby villages of Raduša and Jurumleri with a monthly frequency measuring for physical-chemical parameters of water. The Sampling was implemented according to the ISO 5667. Water samples were collected in polyethylene bottles previously washed with hydrochloric acid and rinsed out with abundant-distilled water in the laboratory and with abundant river water before collection. On the field are measured temperature, pH, conductivity, dissolved oxigen (DO), saturation with WTW Multi 340I set, alkalinity is determined by titration, oxygen fixation was made immediately and measured by Winkler method, biochemical oxygen demand (BOD<sub>5</sub>)and chemical oxygen demand (COD) by KMnO<sub>4</sub>. Other parameters were analyzed in the laboratory of the hydrometeorological service. Hardness by EDTA titration, nutrients is determined by spectrophotometry using UV-VIS Varian Carry 100. Sulfate, chloride, Ca, and Mg by titration. Heavy metals concentration of: Fe, Mn, Pb, Cd, Zn, Cr, Ni, and Cu are determined by atomic absorption spectrometry using Varian 220 by graphite furnace and flame technique. As methods for determination of these parameters were used methods recommended by Standard Methods for the Examination of Water and Wastewater APHA, EPA, and EN/ISO methods. Water quality assessment was based on Macedonian regulation for classification of water quality. According to the Macedonian regulation [10] water was classified in five classes, from very good (class 1) to very bad (class 5).

#### **RESULTS AND DISCUSSION**

In the group of indicators of the basic physical and chemical analysis were taken into account 11 parameters of the two measuring points such as: color (Pt-Co Units), turbidity (NTU), pH, suspended particulars, ammonia (NH<sub>4</sub> <sup>+</sup>), nitrates (NO<sub>3</sub><sup>-</sup>), phosphates (PO<sub>4</sub><sup>-</sup>), nitrites (NO<sub>2</sub><sup>-</sup>), dissolved oxygen, biochemical oxygen demand (BOD) and chemical oxygen demand (COD) from KMnO<sub>4</sub>.

Period	d PARAMETERS										
	Col. (Pt-Co Units)*	Turbidity (NTU)*	pH*	Suspended particulars	NH4 <sup>+</sup>	NO <sub>3</sub> -	PO <sub>4</sub> -	NO <sub>2</sub> -	<b>O</b> <sub>2</sub>	BOD	COD
09/09	8,80	16,00	7,97	18,00	0,13	1,18	0,93	0,08	9,89	3,24	8,02
10/09	6,30	56,00	7,87	58,00	0,02	1,76	0,17	0,05	10,31	4,70	22,76
11/09	3,70	20,00	7,74	21,00	0,12	1,23	0,13	0,03	10,46	2,38	6,16
12/09	4,10	9,00	7,73	11,00	0,11	1,33	0,10	0,02	11,27	2,76	6,72
01/10	5,30	12,00	7,68	13,00	0,07	1,09	0,07	0,01	11,34	0,97	8,65
02/10	5,00	18,00	7,67	19,00	0,08	1,48	0,07	0,01	11,59	1,87	8,57

Table 1. Physico-chemical analysis of Vardar River near the village
Radusha expressed in mg/l, except (*)

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03/10	6,70	50,00	7,79	33,00	0,08	1,28	0,20	0,11	10,93	4,14	6,72
04/10	6,50	22,00	7,73	23,00	0,07	0,81	0,07	0,01	10,76	2,17	8,53
05/10	7,30	30,00	7,73	31,00	0,09	1,05	0,05	0,01	13,79	5,38	7,35
06/10	4,70	17,00	7,69	18,00	0,04	1,31	0,07	0,02	9,73	2,20	12,33
07/10	5,70	21,00	7,74	22,00	0,11	1,31	0,22	0,05	9,38	2,75	9,48
08/10	3,8	17	7,78	19	0,08	3,05	0,12	0,04	9,6	2,11	6,28
Σ	5,66	24,00	7,76	23,83	0,08	1,41	0,18	0,04	10,75	2,89	9,30
(average)											

# Table 2. Physico-chemical analysis of Vardar River near the village Jurumleri expressed in mg/l, except (\*)

Period		PARAMETERS											
	Col. (Pt-Co Units)*	Turbidity (NTU)*	рН*	Suspended particulars	NH4 <sup>+</sup>	NO <sub>3</sub> -	PO <sub>4</sub> -	NO <sub>2</sub> -	<b>O</b> <sub>2</sub>	BOD	COD		
09/09	2,20	21,00	7,50	23,00	2,67	1,60		0,12	3,84	13,8	17,31		
10/09	13,20	78,00	7,50	79,00	0,04	1,33	5,79	0,06	7,79	15,11	43,14		
11/09	5,00	19,00	7,80	20,00	0,36	1,19	0,79	0,03	9,69	6,07	7,47		
12/09	9,00	25,00	7,80	28,00	0,99	1,8	0,29	0,04	9,89	8,36	10,63		
01/10	6,20	15,00	7,80	17,00	0,3	1,09	0,19	0,01	11,26	4,79	9,21		
02/10	6,80	25,00	7,80	27,00	0,33	1,55	0,03	0,02	11,06	7,04	9,91		
03/10	6,40	34,00	7,70	35,00	0,35	1,24	0,29	0,03	10,72	7,14	14,42		
04/10	8,60	42,00	7,70	44,00	0,28	1,42	0,03	0,02	9,89	4,41	12,64		
05/10	6,70	13,00	7,40	15,00	0,5	0,96	0,22	0,03	12,12	4,49	5,77		
06/10	8,00	20,00	7,50	22,00	0,46	1,47	0,22	0,05	8,97	4,98	13,91		
07/10	0,00	9,00	7,50	10,00	0,44	1,77	0,26	0,07	8,18	5,92	10,51		
08/10	4,20	5,00	7,36	6,00	0,30	1,19	0,25	0,02	11,81	5,79	6,36		
Σ (average)	6,36	25,50	7,61	27,17	0,59	1,38	0,76	0,04	9,60	7,33	13,44		

The color as an indicator of the two measuring points in the river Vardar in the investigated period shows constant values of quality Class I, and apparently in the lower measuring point at the village Jurumleri there is minimal deterioration in terms of annual averages (tab. 1, 2).

Turbidity as a form of mechanical water pollution directly affects the dispersion of light, oxygen saturation, photosynthesis and photolysis [7]. Samples of the two measuring points demonstrate permanent turbidity of the water of the river Vardar throughout the year with values appropriate of quality class V. The poor quality of water present in the upper measuring point at the village Radusha (24.0) is further exacerbated at the lowest point (25,5), in. Jurumleri (tab. 1, 2).

The acidity (pH) in all samples throughout the year from the two measuring points correspond to a quality class I with some increase in acidity downstream. (tab. 1, 2). Optimal range of pH in rivers exponent is ranging from 6,5-8,0 [1]. The low pH value of the exponent,

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or the increased acidity causes the dissolution of toxic metals and is a fatal threat to the organic world [6].

Suspended solids, as an indication of mineralization, are usually the result of natural erosion processes. In terms of this indicator we see that the river Vardar in Skopje Valley enters with quality class II near the village Radusha averaging 23, 83 mg/l and the same quality class retains the lower measuring point at the village Jurumleri with a slight increase in the concentration of an above 27,12 mg/l (tab. 1, 2).

From the forms of nitrogen as a nutrient, ammonium ion (NH4<sup>+</sup>) is toxic and unstable form which elevated concentrations reflect the burden of aquatic ecosystems waters, especially if the amount of dissolved oxygen is low. At the measuring point in the village Radusha that refers to concentrations of quality class I-II (average 0, 08 mg/l), and the impact of the sewage from the city of Skopje and other villages on the waters of Vardar is visible on the lower measuring point at the village Jurumleri, where the concentrations of ammonium ion increased to seven times than the average 0, 59 mg/l (tab. 1, 2). The input of nitrates (NO<sub>3</sub><sup>-</sup>) as a form of nitrogen in the river water goes through disposal of animal waste, feces and washing of soil treated with fertilizers [12]. The concentration of nitrates in the Vardar River in Skopje Valley through the examined period is held within quality class I-II (tab. 1, 2). Here we record and some decrease from the top (1,41 mg/l) to the lowest measuring point (1,38 mg/l). Nitrite (NO<sub>2</sub>) is an intermediate product between ammonia and nitrates. Nitrite is a toxic form of nitrogen for the wildlife in waters but in very low concentrations. In the waters of Vardar in Skopje Valley at the two measuring points were recorded annual concentrations of 0,04 mg/l, corresponding quality class III-IV and undoubtedly includes anthropogenic factor for their input through wastewater, agricultural and industrial activities.

Phosphorus as a nutrient was tested through orthophosphate ion (PO<sub>4</sub><sup>-</sup>). It comes from the minerals of the lithosphere and the fertilizers. It is non-toxic to living organisms in the water so that it in the legal norms of the Republic of Macedonia there are no prescribed limits. However, its annual monitoring in the waters of Vardar and obvious increase from the measuring point in Radusha (0,18 mg/l) to that of Jurumleri (0,76 mg/l) shows that the treatment in this area with fertilizers in agricultural activities is accurate.

Among the forms of oxygen regimes, the subject of monitoring were: dissolved oxygen, biochemical oxygen demand and chemical oxygen demand. Dissolved oxygen is an indicator of "health" of the water ecosystem and the metabolic processes related to the degradation of existing organic matter [9]. According to this indicator the river Vardar in Skopje Valley near the village Radusha getting high quality (Class I) with an average annual amount of 10.75 mg/l. It retains the same class without expressed monthly fluctuations also on the lower measuring point at Jurumleri with average 9, 60 mg/l. The anthropogenic impact on the water quality of Vardar in the Skopje Valley is visible through biochemical oxygen demand (BOD) which indicates the presence of organic matter decomposition that's enhanced consumes

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dissolved oxygen. The measuring point in Radusha (point 1) indicates an average annual quality class II (2,89 mg/l), while at the lower measuring point in Jurumleri (point 2) the quality drops to class IV (7,33 mg/l). Chemical oxygen demand (COD) by KMnO<sub>4</sub> is an indicator that reflects oxygen consumption for total decomposition and oxidation process of the present metals in the water. Deterioration in the quality of Skopje section is obvious (tab. 1, 2), and also the involvement of anthropogenic factor, so that in the upper control point shows average quality of class II (9, 30 mg/l), and at the lower point shows quality of class IV (13, 44 mg/l). Seasonal, the situation is most critical during the summer months due to reduced water levels and the increased concentration of organic materials and metals.

Regarding the burden of the Vardar River water with heavy metals, reviewed were: iron, manganese, lead, zinc, chromium, nickel, and copper (tab. 3, 4). These substances are considered toxic in relatively low concentrations to plant and animal life and tend to accumulate in the food chain.

Period	PARAMETERS										
	Fe	Mn	Pb	Zn	Cd	Cr	Ni	Cu			
09/09	15,00	33,00	1,18	6,00	0,52	3,25	0,25	1,43			
10/09	60,00	28,00	0,48	28,00	0,30	1,55	0,60	9,40			
11/09	33,00	10,00	1,19	0,20	0,01	3,40	1,40	0,42			
12/09	27,00	17,00	0,49	7,00	0,00	5,88	0,47	0,05			
01/10	58,00	2,00	1,91	23,00	0,10	5,89	5,15	3,14			
02/10	53,00	18,00	1,47	23,00	0,04	2,61	1,34	2,81			
03/10	111,00	6,00	0,31	22,00	0,10	2,94	6,00	4,14			
04/10	111,00	6,00	0,83	21,80	0,03	7,46	0,03	2,62			
05/10	60,00	14,00	3,92	25,00	0,11	7,70	3,10	1,61			
06/10	52,00	3,00	0,89	14,00	0,08	13,45	0,55	2,52			
07/10	69,00	11,00	1,44	32,00	0,10	18,50	4,83	1,78			
Σ (average)	59,00	13,45	1,28	18,36	0,13	6,60	2,16	2,72			

#### Table 3. Concentrations of heavy metals in the water of Vardar River near the village Radusha (expressed in μg/l)

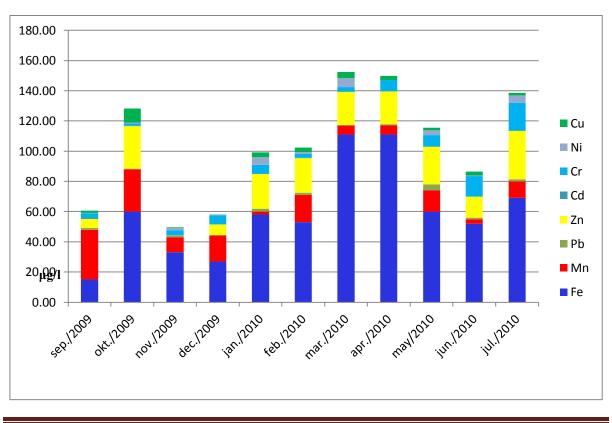
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Period	PARAMETERS										
	Fe	Mn	Pb	Zn	Cd	Cr	Ni	Cu			
09/09	21,00	46,00	2,85	6,00	0,23	0,07	1,91	1,50			
10/09	226,00	66,00	0,72	52,00	0,72	1,64	4,19	21,84			
11/09	73,00	2,00	2,53	13,60	0,10	1,39	0,52	1,86			
12/09	69,00	29,00	2,76	51,00	0,05	9,60	0,93	0,14			
01/10	529,00	9,00	1,33	46,00	0,07	2,46	4,36	3,70			
02/10	167,00	15,00	1,66	0,06	0,09	2,19	0,51	5,61			
03/10	95,00	19,00	0,74	44,00	0,09	1,92	1,85	4,29			
04/10	95,00	19,00	3,25	43,70	0,13	2,86	1,96	6,31			
05/10	169,00	1,00	1,33	35,00	0,14	2,11	0,28	3,45			
06/10	133,00	2,00	0,25	55,00	0,04	5,80	1,20	2,78			
07/10	40,00	40,00	2,51	61,00	0,06	9,58	4,99	0,56			
2 (average)	147,00	22,55	1,81	37,03	0,16	3,60	2,06	4,73			

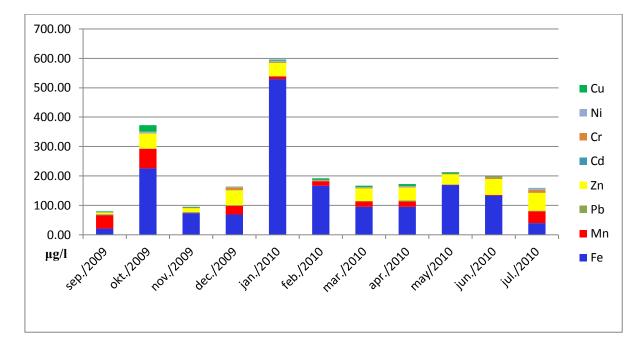
## Table 4. Concentrations of heavy metals in the water of the river Vardar near the village Jurumleri (expressed in µg/l)

#### Figure 2. Annual regime of the concentration of heavy metals in the water of the river Vardar near the village Radusha (expressed in µg/l)



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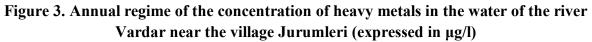
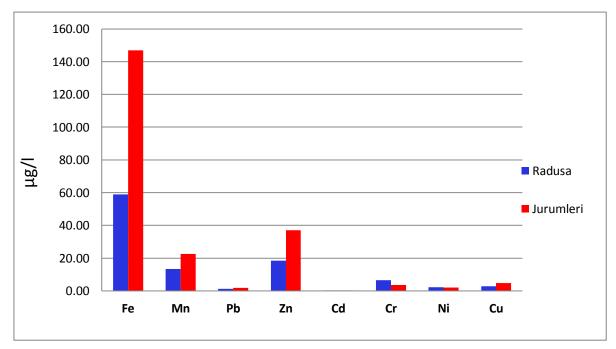


Figure 4. Average annual values of concentration of heavy metals in the water of the river Vardar near the village Radusha and Jurumleri



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Iron is one of the most widespread metals in the lithosphere and surface waters. The increased concentration of iron in the water is distracting the osmo regulation of the plant, and thus the quality of agricultural food. In irrigation water it is not recommended to be with concentrations above 5  $\mu$ g/l [4]. In the waters of river Vardar in Skopje Valley in the upper measuring point the average year concentrations correspond to quality of Class I (59,0  $\mu$ g/l) and in the lower measuring point with the same quality but with elevated values of 147, 0  $\mu$ g/l (tab. 3, 4; fig. 2, 3, 4). Dangerous concentrations of iron were detected only in January in Jurumleri (point 2, fig. 1) with 529  $\mu$ g/l, referring to quality of Class III-IV, which reflect the tested anthropogenic influence of the section between the two measuring points.

Manganese in water is naturally derived mostly from dissolution of shales and sandstones, and throws the man through metallurgical activities. That reduces photosynthesis in plants by reducing the absorption of sunlight. In irrigation water it must not exceed a concentration of  $0,2 \ \mu g/l$  [4]. At the two measuring points in Skopje Valley in the water of the Vardar River that concentration has dramatically exceeded, but under national rules of classification still remains within the class I. The increase in concentration occurs downstream from 13, 45  $\mu g/l$  to 22, 55  $\mu g/l$  (tab. 3, 4; fig. 2, 3, 4).

Lead is a heavy metal which in the rivers can be deposited by mining and industrial waste from factories, military assets, batteries, etc [7]. Irrigation water in its concentration, in a long-term, should not exceed 5,0  $\mu$ g/l [5]. Lead as a toxic metal is accumulated in the roots of plants. The clearness of the river water through Vardar Skopje Valley for agricultural needs is quite high from the average values examined period at the two measuring points (1,28  $\mu$ g/l and 1,81  $\mu$ g/l) which constantly keeps the water quality within the class I-II (tab. 3, 4; fig. 2, 3, 4).

Zinc is an essential element for plants and animals as it is necessary for the functioning of certain enzymes. Zinc is relatively non-toxic to terrestrial organisms. It is acutely and chronically toxic to aquatic organisms, particularly fish. Zinc toxicity decreases with increasing hardness, increases with increasing temperature, and increases with decreasing dissolved oxygen [6]. If it is known that in unpolluted water its concentration does not exceed 5  $\mu$ g/l, then the visible impact of anthropogenic factor on the waters of Vardar River in Skopje Valley where even within the class I-II, water quality downstream deteriorate at an average from 18,36  $\mu$ g/l (Radusa) to 37,03  $\mu$ g/l (Jurumleri).

Cadmium is a toxic metal which is very rare in nature. In some rivers in R. Macedonia is naturally present originating in certain Paleozoic and Mesozoic rocks [8]. The human factor introduces cadmium into the waters by cement dust, which is present in Skopje Valley, thorough melting of zinc ore, production of batteries and more. In irrigation water must not exceed 0,01  $\mu$ g/l in long terms, because it performs a reduction in plant growth and changes in cell metabolism by reduced chlorophyll [11]. The presence of cadmium in the waters of Vardar in Skopje Valley at the two measuring points in the investigated period on average

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slightly exceeded the maximum permissible limits and reports values of quality class III-IV (tab. 3, 4; fig. 2, 3, 4). The slight increase in its concentration of 0,13  $\mu$ g/l (Radusa) to 0,16  $\mu$ g/l (Jurumleri) may be due to the industrial impact (cement industry, etc.).

Chromium is a heavy metal naturally present in chromite ore (FeCr<sub>2</sub>O<sub>4</sub>). The content of his toxic form in hexavalent state (Cr<sub>6</sub><sup>+</sup>) in the waters of Vardar in Skopje Valley is very interesting considering the fact that near the upper measuring point (Radusa) there is an abandoned chromium mine and ferrochrome alloy smelting plant in the village Jegunovce. Its concentration in irrigation water must not exceed 0,1  $\mu$ g/l on long term periods [5]. That is why during the summer months during a lower flow above the measuring point (point 1., fig. 1) were observed concentrations appropriate to quality class III-IV, while at the lower control point (point 2., fig. 1.) the values of the hexavalent chromium in the Vardar River constantly maintained within the class I-II, with an average of 3, 60  $\mu$ g/l (tab. 3, 4; fig. 2, 3, 4).

Nickel is the companion of iron in the mineral magnetite, and its emissions in water may be the result of mining waste, factories for batteries and nickel products. His concentration over Vardar Skopje valley in the examined period consistently maintained low levels of both measuring points (2,16  $\mu$ g/l and 2.06  $\mu$ g/l) which corresponds to quality of class I-II (tab. 3, 4 ; fig. 2, 3, 4) excluding the anthropogenic factor for its presence.

Copper as a heavy metal in aquatic ecosystems is derived from certain minerals (limestone, sandstone), mining and metallurgical activities and excessive use of fungicides in agriculture [9, 14]. In the irrigation water it must not exceed a concentration of 0, 1  $\mu$ g/l. In the waters of Vardar River in Skopje Valley its concentration on average doubles, which emphasizes the anthropogenic impact, although at the two measuring points the average values correspond to quality of class I-II with only few monthly outages to class III-IV at the lower measuring position (tab. 3, 4; fig. 2, 3, 4).

#### CONCLUSION

In this paper were studied the waters of Vardar River in Skopje Valley which were monitored at two measuring points: at the entrance to the valley near the village Radusha (point 1, fig. 1) and downstream nearby Jurumleri (point 2, fig. 1). The assessment of the water quality of Vardar was based on 11 parameters of the basic physical and chemical analysis and 8 heavy metals as a criterion for quality that have a significant impact on agricultural production during irrigation conditions. In terms of basic physico-chemical analysis it was determined the deterioration of water quality around Skopje Valley compared to 10 parameters, except for nitrates. Best water quality was observed in terms of color, pH exponent, ammonium ion, dissolved oxygen and nitrates (Class I-II) of the two measuring points. Lowest water quality was registered in terms of turbidity (class V), BOD (class IV) and COD (Class IV) and nitrite (class III-IV) at the lower measuring point, which undoubtedly highlights the organic load of the Vardar River from anthropogenic sources of pollution, thorough the input of sewage,

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industrial waste and excessive use of chemicals in agriculture. Regarding the presence of heavy metals in the waters of Vardar River through Skopje area, was established deterioration of the quality based on 6 indicators (Fe, Mn, Pb, Zn, Cd, and Cu) although most express concentrations within the class I-II, except cadmium which is present at concentrations of class III-IV. Reducing the concentration downstream is noticed only in terms of two parameters (Ni, Cr). The annual regime of heavy metals in the water, although insignificant but still reflects the industrial pollutants that are discharged into the Vardar River in Skopje Valley, and we must take strict preventive measures about it. Therefore, the use value of waterflow under conditions of irrigation is reduced relative to the 5 indicators of heavy metals (Fe, Mn, Cd, Cr, Cu) are showing exceeded concentrations for environmental correctness of the irrigational waters.

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