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Assessment of water pollution of the South Morava River (Serbia) by statistical and index methods

ABSTRACT

Factor analysis/principal component analysis (FA/PCA), applied to 16 physico-chemical parameters at three different monitoring sites (Mojsinje, Korvingrad, and Ristovac) of the South Morava River in 2015, and from 2011-2018 for station Ristovac, extracted two and four varimax factors which explain 100.000 and 90.874 % of the total variance in water quality, respectively. The relationships among the stations and years obtained by cluster analysis (CA) categorize observed objects in different quality levels. Parameters responsible for pollution of the South Morava River are related to point (industrial/sewage effluents), non-point (runoff from arable land and erosion), and natural source (a mineral component of the river water). The results of the water pollution index (WPI) method for 2015 have shown that the South Morava River at Korvingrad station represents a "moderately polluted" water body (class III), but at Mojsinje and Ristovac station a "polluted" water body (class IV). The obtained ecological potential values indicated that water quality at the Korvingrad station corresponds to class II, "good ecological potential". The surface water at Mojsinje and Ristovac stations deviated from the required ecological water standards by the European Union Water Framework Directive (EU WFD), class III. The South Morava River had the highest load of nutrients (PO_4 -P and TP) and organic matter (TOC) at Ristovac station, which was confirmed by the index and comparative method (polluted water with poor ecological potential) in period from 2011-2018. A comprehensive analysis of the paper identified a decreasing trend in pollution of the South Morava River, primarily at Ristovac station.

Keywords: South Morava River, water quality, multivariate statistical techniques, water pollution index, ecological status/potential

1. INTRODUCTION

Surface waters must be sustainably used and protected because they affect human health and aquatic ecosystems. Also, surface waters have an indirect impact on agriculture, industry, transport, and water supply [1,2]. The quality of watercourses depends on several factors: geological (lithology of the basin), erosion, climatic conditions, and anthropogenic factor [3,4]. Human activities become significant variables (urbanization, agricultural development, land drainage, and discharge of pollutants) which determine the quality of surface water [4-6]. The surface water quality is expressed through physical, chemical, and biological parameters [7].

The river represents a vulnerable type of water system to pollution, due to the possibility of inflow of municipal and industrial wastewater and the possibility of easy disposal of waste on its banks [8,9]. Atmospheric precipitation also contributes to pollution through runoff from agricultural land and surface runoff [5,10]. In order to effectively manage rivers, it is necessary to control and monitor the pollution of watercourses from point/non-point sources in the long term [11].

In the assessment of surface water quality, complex data sets of quality indicators are obtained, which needs to be simplified to identify the type of pollution. Interpretation of a large number of parameters and their variability by multivariate methods, such as factor analysis/principal component analysis (FA/PCA) and cluster analysis (CA), allows identification of the influence of pollution sources on aquatic systems and providing reliable information on degradation of water quality and ecological status. In this way,

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efficient management of water resources is being established [9,12-15]. These methods are an appropriate tool for determining spatial and temporal variations in the quality of water as well as for separating monitoring stations of similar or different quality [16,17].

In addition to statistical methods, the ecological significance of surface waters is estimated by mathematical methods which define quality indicators and determine ecological status [18-20]. The European Union (EU) Water Framework Directive (Directive 2000/60/EC - WFD) defines water quality standards and requires the achievement of good ecological and chemical status of surface waters, which significantly reduces surface water pollution and creates a policy of management and monitoring of river systems. Each EU member state adapts its water laws and regulations with EU water legislation [21-23]. The water pollution index (WPI) is one of the mathematical indexes that is increasingly used in water pollution analysis. It usefully summarizes a large amount of the physical, chemical, and biological water quality data in a single value determining degrees of water pollution [22]. The WPI index not only represents the classification of water systems based on measured parameters but also indicates the type of pollution in a unique way, i.e. potential contaminants of water ecosystems [18].

During the last decade, Serbia has been focusing significantly on the protection and preservation of water resources and definition of environmental monitoring system for surface- and groundwater. On the rivers, many monitoring stations have been established, which gives a large number of measurements indicating a disproportion between the required and current state of water quality [23,24]. In the works of other authors, the water quality of the South Morava River (part of the Danube River system) in the period from 2005 to 2012 was analyzed [4,25]. The South Morava River is the recipient of various pollutants, and the water at the Ristovac monitoring station (the entry profile of the South Morava River) is under the heaviest pollutant loads. Special attention must be given to the assessment of the water quality of the South Morava River based on recent physical and chemical data.

The present study applied a new approach to the analysis of water quality obtained by combining multivariate statistical and index methods, which gave results on the pollution degree of the South Morava River. Data sets obtained during the monitoring period from 2011 to 2018 were processed by statistical methods to identify water

quality variables and determine differences between monitoring stations and the monitoring periods. The analysis was completed by determining the class of ecological status/potential and their deviations from the requirements of the EU WFD.

2. EXPERIMENTAL

2.1. Study Area

The South Morava River is located in the southeast of Serbia and is formed by the confluence of the Binačka and Preševska Moravica near Bujanovac town. It flows in a south-north direction towards central Serbia and forms the Great Morava with the West Morava. The river belongs to the Black Sea basin with 295 km length and a basin area of 15496 km² (85% belongs to Serbia). The South Morava basin has a hilly and mountainous character, in the range of altitudes from 300 to 2169 m, and the river valley which is composed of a large number of gorges and depressions. The most important left tributaries of this river were the Jablanica, the Veternica, the Pusta Reka, and the Toplica, and from the right tributaries: the Vrla, the Vlasina, the Sokobanjska Moravica, and the Nišava (the longest tributary) [25-27].

The South Morava River is a partially utilized resource for electricity production and irrigation [25]. The biggest polluters of the South Morava River are the industries in Preševo, Bujanovac, Vranje, Vladičin Han, Leskovac, and Niš town [26]. The deterioration of river water quality is caused by bank erosion and the arrival of contaminants into the watercourse, as well as the inflow of municipal and industrial wastewaters [27-29].

Three monitoring stations were selected for the analysis of water quality of the South Morava River, with a distance from the mouth into the Great Morava: Ristovac (entry monitoring profile) 237.0 km, Korvingrad 105.7 km, and Mojsinje (exit monitoring profile) 16.4 km (Figure 1).

2.2. Materials and methods

The research is based on the data from the Environmental Protection Agency, for the period from 2011 to 2018 (Ministry of Environmental Protection of the Republic of Serbia 2011-2018) [30] for 16 physico-chemical parameters: turbidity (T), suspended solids (SS), dissolved oxygen (DO), pH, electrical conductivity (EC), bicarbonates (HCO₃⁻), calcium (Ca²⁺), magnesium (Mg²⁺), chloride (Cl⁻), sulphate (SO₄²⁻), ammonium ion (NH₄-N), nitrate (NO₃-N), orthophosphate (PO₄-P), total phosphorus (TP), 5-days biochemical oxygen demand (BOD₅) and total organic carbon (TOC).

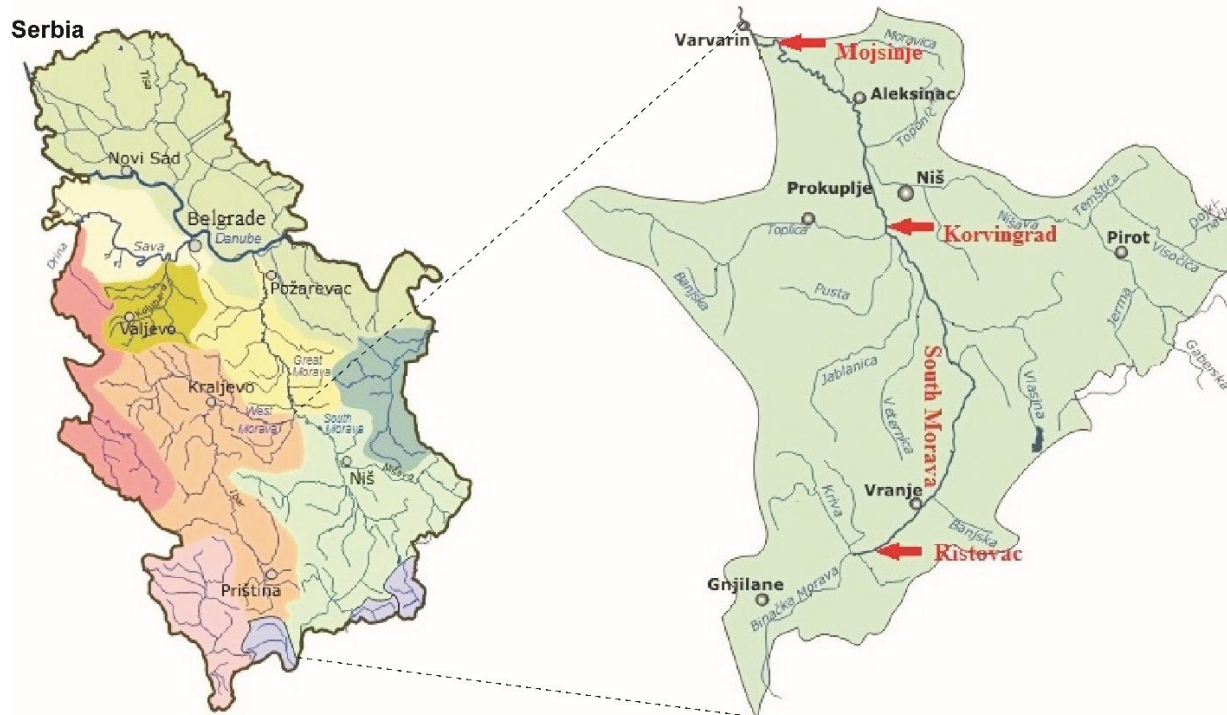


Figure. 1 Monitoring stations at the South Morava River in the Republic of Serbia

Slika 1. Monitoring stanice na reci Južna Morava u Republici Srbiji

A one-month sampling of surface water was performed at monitoring stations and physico-chemical parameters were determined in certified laboratories by relevant methods: T - UP 1.88/PC 12, SS - APHA AWWA&WEF, part 2540 D:2005, DO - UP 1.89/PC 12, pH - SRPS H.Z1.111:1987, EC - UP 1.95/PC 12, HCO_3^- - SRPS EN ISO 9963-1:2007, Ca^{2+} - ISO 6058:1984, Mg^{2+} - ISO 6059:1984, Cl^- - SRPS ISO 9297:1997, SO_4^{2-} - UP 1.101/PC 12, $\text{NH}_4\text{-N}$ - UP 1.96/PC 12, $\text{NO}_3\text{-N}$ - UP 1.98/PC 12, $\text{PO}_4\text{-P}$ - UP 1.102/PC 12, TP - APHA AWWA WEF 4500 (A, B, E), BOD_5 - UP 1.4/PC 12 and TOC - SRPS ISO 8245:2007.

The EU WFD defines reference conditions (undisturbed water system, which suffers from very low anthropogenic pressure) and sets requirements for monitoring surface- and groundwater. The directive includes an analysis of the current status of surface water and the impact of human activity on water system quality (until 2024) and taking measures to achieve a "good eco-chemical status" of the watercourses. In Serbia, this concept of WFD has been implemented through the enactment of the Regulation on determining the water bodies of surface water and groundwater (Official Gazette of the Republic of Serbia, No. 96/2010) [31], the Regulation on the parameters of ecological and chemical status of surface water and parameters of the chemical and quantitative status of groundwater (Official Gazette of the

Republic of Serbia, No. 74/2011) [32], and the Regulation on reference conditions of surface water types (Official Gazette of the Republic of Serbia, No. 67/2011) [33]. The ecological status/potential of surface waters is defined by the elements of biological, physico-chemical, and hydromorphological quality for a certain category and type of water body. The ecological status of rivers and lakes is classified into five classes: as excellent (class I), good (class II), moderate (class III), poor (class IV), and bad (class V), and the ecological potential of heavily modified and artificial water bodies also in 5 classes: as maximum (I), good (II), moderate (III) poor (IV), and bad (class V). Heavily modified and artificial water bodies, specific categories of water bodies, are watercourse sectors that have undergone significant changes under the influence of human activities which can achieve "good ecological potential" but not "good ecological status".

According to the Regulation on determining the water bodies of surface water and groundwater (Official Gazette of the Republic of Serbia, No. 96/2010) and Regulation on the parameters of ecological and chemical status of surface water and parameters of the chemical and quantitative status of groundwater (Official Gazette of the Republic of Serbia, No. 74/2011), the South Morava River is classified into type 2 watercourses, large rivers with a dominance of medium sediment,

except for the rivers of the Pannonian Plain. The watercourse South Morava is classified as a river at the monitoring station Mojsinje, while at the monitoring stations Korvingrad and Ristovac it is

categorized as a heavily modified water body. Table 1 gives the limit values of physico-chemical parameters for determining the ecological status/potential for a water body of type 2.

Table 1. Physico-chemical limits of ecological status and potential for water bodies type 2 [32]

Tabela 1. Fizičko-hemijske granice ekološkog statusa i potencijala za vodno telo tipa 2 [32]

Parameter		Limits between the classes of ecological status				Limits between the classes of ecological potential		
		I-II	II-III	III-IV	IV-V	II-III	III-IV	IV-V
pH	-	6.50-8.50	6.50-8.50	6.50-8.50	<6.50; >8.50	6.50-8.50	6.50-8.50	<6.50; >8.50
DO	mg/L	8.50	7.00	5.00	4.00	7.00	5.00	4.00
NH ₄ -N	mg/L	0.05	0.10	0.80	1.00	0.10	0.80	1.00
NO ₃ -N	mg/L	1.50	3.00	6.00	15.00	3.00	6.00	15.00
PO ₄ -P	mg/L	0.02	0.10	0.20	0.50	0.10	0.20	0.50
TP	mg/L	0.05	0.20	0.40	1.00	0.20	0.40	1.0
BOD ₅	mg/L	1.80	4.50	6.00	20.00	4.50	6.00	20.00
TOC	mg/L	2.00	5.00	7.00	23.00	5.00	7.00	23.00
Cl ⁻	mg/L	50.00	100.00	/	/	50.00	100.00	/

In the paper, ecological status/potential are determined by comparing the average annual concentrations of physico-chemical parameters with their concentration limits for the appropriate class prescribed by Regulation on the parameters of ecological and chemical status of surface water and parameters of the chemical and quantitative status of groundwater (Official Gazette of the Republic of Serbia, No. 74/2011), Table 1.

Water pollution index (WPI) is an arithmetic way of integrating many different parameters, mostly physical and chemical, to evaluate the surface water status [22,34]. The WPI index method is based on the determination of the deviation degree of a specific parameter from the allowed values by regulative, which results in the identification of factors that have a significant influence on pollution. Also, the WPI method allows comparative analysis of water quality of two or more watercourses [18].

WPI is calculated as the sum of ratios between the observed parameters (average annual concentration of analyzed parameters - C_i) and prescribed standard threshold values by regulative (SFQS), divided by the number of used parameters (n) [22,35]:

$$WPI = \sum_{i=1}^n \frac{C_i}{SFQS} \times \frac{1}{n} \quad (1)$$

Standard threshold values of parameters are defined for each country by regulations, and in Serbia, SFQS are values of parameters for class I of surface water according to national legislation

(Official Gazette of the Republic of Serbia, No. 74/2011) [23,35].

The WPI value is presented through 6 water classes (Table 2) according to the requirements of the EU WFD and indicates the ecological status/potential. Surface waters according to the WPI index are classified from "very pure" ($WPI \leq 0.3$) to "heavily impure" ($WPI > 6$) water bodies. The index is expressed as a single value that is comparable to other index systems in Europe [22,34].

Table 2. Surface water classification based on WPI index [34]

Tabela 2. Klasifikacija površinskih voda bazirana na WPI indeksu [34]

Class	Characteristics	WPI
I	Very pure	≤ 0.3
II	Pure	0.3-1
III	Moderately polluted	1-2
IV	Polluted	2-4
V	Impure	4-6
VI	Heavily impure	>6

Multivariate statistical methods were used for the identification of the factors responsible for the water quality variability of the South Morava River (factor analysis/principal component analysis - FA/PCA) as well as for hierarchical clustering monitoring stations/years based on detecting similarities and differences between physico-chemical water quality parameters (cluster analysis - CA).

Principal component analysis (PCA) is a technique of obtaining new uncorrelated variables (principal components - PC) by a linear combination of original variables [36]. This interdependence technique is used to identify the most important parameters that describe water quality variability without losing the original information data [37]. The principal component (PC) can be expressed as [38]:

$$z_{ij} = a_{i1}x_{1j} + a_{i2}x_{2j} + a_{i3}x_{3j} + \dots + a_{im}x_{mj} \quad (2)$$

where z is the component score, a is the component loading, x is the measured value of variable, i is the component number, j is the sample number, and m is the total number of variables.

Factor loading values obtained from principal component analysis do not always explain the initial data sets. Rotation of factors (i.e. PCA) was extracted varimax factors (VF), which contain less significant variables and allow easier interpretation of a complex data matrix [9,37]. The described factor analysis can be represented by the following expression [38]:

$$z_{ji} = a_{f1}f_{1i} + a_{f2}f_{2i} + a_{f3}f_{3i} + \dots + a_{fm}f_{mi} + e_{fi} \quad (3)$$

where z is the component score, a is the component loading, f is the factor score, e is the residual term accounting for errors or other source of variation, i is the sample number and m is the total number of variables.

The eigenvalue greater than 1 (Kaiser's criterion) was taken as a criterion for the determining of a number of principal components in the FA/PCA analysis. The orthogonal rotation method (Varimax) was used to obtain factors that enabled adequate water analysis in a function of target parameters.

Cluster analysis or grouping analysis (CA) is a method by which objects are grouped into clusters based on similarity according to a predetermined criterion. Hierarchical CA is one of the most commonly used approaches that allows determining the relationship between parameters and the entire data set based on similarity [9,39]. In the paper, hierarchical agglomerative cluster analysis (HACA) was performed on the data set by the Single Linkage method. The distance between all objects (monitoring stations and years) was determined using the Euclidean distance. An expression for Euclidean distance between two objects, i and j , is:

$$d_{ij} = \left(\sum_{k=1}^m (z_{ik} - z_{jk})^2 \right)^{1/2} \quad (4)$$

Where d_{ij} is the Euclidean distance, z_{ik} and z_{jk} are the values of variable k for object i and j , respectively, and m is the number of variables.

The hierarchical structure of objects is shown by a hierarchical tree (dendrogram). The cluster analysis was performed in relation to the identified varimax factors by FA/PCA method.

For multivariate analysis, the average annual concentrations of quality parameters were used as input data, which were analyzed using the software package Statistica Version 7.0 (Statsoft, Tulsa, Oklahoma, USA).

3. RESULTS AND DISCUSSION

EU WFD deals with creating a strategy for monitoring surface-, ground-, and seawaters [21]. Supervisory and/or operational monitoring of surface water quality in Serbia was performed at around 180 monitoring profiles. The analysis in this paper completes the monitoring system from the research aspect for the selected surface water body, the South Morava River. In this way, the direction of preventive action is defined to signaling, controlling, and reducing the threat to this water body.

Table 3. Average annual values of water quality parameters at monitoring stations of the South Morava River for 2015

Tabela 3. Srednje godišnje vrednosti parametara kvaliteta vode na monitoring stanicama reke Južne Morave za 2015. godinu

Parameter	Unit	Monitoring station		
		Mojsinje	Korvingrad	Ristovac
T	NTU	135.96	38.17	69.71
SS	mg/L	98.25	44.75	63.92
DO	mg/L	10.21	10.92	8.88
pH	-	8.03	8.13	7.84
EC	mS/cm	406.75	378.17	607.92
HCO ₃ ⁻	mg/L	200.25	182.67	305.42
Ca ²⁺	mg/L	53.83	51.00	82.08
Mg ²⁺	mg/L	14.83	13.75	22.58
Cl ⁻	mg/L	11.46	13.47	15.77
SO ₄ ²⁻	mg/L	24.50	27.67	46.25
NH ₄ -N	mg/L	0.11	0.10	0.14
NO ₃ -N	mg/L	1.28	1.22	1.77
PO ₄ -P	mg/L	0.086	0.079	0.108
TP	mg/L	0.304	0.199	0.294
BOD ₅	mg/L	3.37	2.76	3.57
TOC	mg/L	5.60	4.79	5.76

The average annual values of the defined water quality parameters at the monitoring stations (Mojsinje, Korvingrad, and Ristovac) of the South Morava River in 2015 are shown in Table 3. The Environmental Protection Agency carried out measurements at all three monitoring stations at the South Morava River the last time in 2015, and

therefore this year was chosen for analysis in order to obtain the most authoritative data of water quality.

From the values of quality indicators, which were estimated for 2015 (Table 3), fluctuations in turbidity (T) and suspended solids (SS) were observed. The highest T and SS values were recorded at the monitoring station Mojsinje 135.96 NTU and 98.25 mg/L, respectively. The water of the South Morava River has a basic character, whereby pH values differ slightly between stations. The oxygen regime is relatively uniform at the monitoring stations Mojsinje and Korvingrad, 10.21 and 10.92 mg/L, while the decrease of oxygen content value occurs at the Ristovac station, 8.88 mg/L (Table 3). According to the indicators of organic pollution (BOD₅ and TOC), nutrients (NO₃-N and PO₄-P), and EC, the Ristovac station stands out from all monitoring stations, so the water in this sector of the watercourse can be considered the most polluted. Higher concentrations of ions in the water of the Ristovac station caused a high water value of EC. The decrease of oxygen content in the water of this station is probably due to the increase in the activity of microorganisms that decompose organic matter and/or the arriving of nutrients that stimulate the growth of algae (eutrophication process) [40].

Table 4. Loadings of a varimax factor for analysed water quality parameters of the South Morava River

Tabela 4. Opterećenja varimaks faktora za analizirane parametre kvaliteta vode reke Južne Morave

Parameter	VF1	VF2
T	-0.070	-0.997
SS	-0.110	-0.994
DO	0.997	0.077
pH	0.997	0.074
EC	-0.987	0.157
HCO ₃ ⁻	-0.990	0.139
Ca ²⁺	-0.982	0.189
Mg ²⁺	-0.987	0.159
Cl ⁻	-0.727	0.687
SO ₄ ²⁻	-0.918	0.396
NH ₄ -N	-0.999	0.030
NO ₃ -N	-0.985	0.172
PO ₄ -P	-0.999	0.039
TP	-0.651	-0.759
BOD ₅	-0.860	-0.510
TOC	-0.813	-0.581
Eigenvalue	12.066	3.934
%Total variance	75.410	24.590
Cumulative % variance	75.410	100.000

The water of the South Morava River at the Mojsinje station is characterized by a high value of T, SS, TP, BOD₅, and TOC, which may indicate a significant impact of nonpoint or point pollution source on the quality of watercourse. In the observed period, the purest water in relation to analyzed parameters was identified at monitoring station Korvingrad.

FA/PCA analysis was done based on average values of parameters for three monitoring stations, Mojsinje, Korvingrad, and Ristovac (Table 3), and FA/PCA results are given in Table 4.

Factor loadings according to Liu et al. [41] indicate "strong" (>0.75), "moderate" (0.75-0.50), and "weak" correlation (0.50-0.30) between quality parameters and extracted factor. The "strong" and "moderate" factor loadings are considered significant for FA/PCA analysis.

Two varimax factors explain 100% of the total variance of river water quality. The VF1 factor accounted for 75.41% in total variance, and it is strong positively associated with DO and pH and strong negatively associated with EC, HCO₃⁻, Ca²⁺, Mg²⁺, SO₄²⁻, NH₄-N, NO₃-N, PO₄-P, BOD₅, and TOC. Also, it has moderate negatively loaded with Cl⁻ and TP. The factor contains minerals, nutrients, and organic load indicators that originated from sewage/industrial effluents (point pollution source) and runoff from the agricultural land (non-point pollution source). Sewage/industrial effluents are naturally acidic with a higher concentration of Na⁺, K⁺, Cl⁻, SO₄²⁻, NH₄-N, and total dissolved solids, which in turn decreases the pH [42]. The presence of degradable organic matter was reflected by a decrease in oxygen concentration in the water.

The VF2 factor accounted for 24.59% of the total variance and was correlated with the following parameters: T, SS, Cl⁻, TP, BOD₅, and TOC. It is strong negatively associated with T, SS, and TP, while moderate negatively associated with BOD₅ and TOC and represents a factor of bank erosion and terrain leaching. The erosion of the banks on which is deposited waste, during the wet period, results in the presence of suspended particles, phosphorus, and organic matter in the water. Phosphorus is present in larger quantities in the soil after the application of fertilizers.

Due to the high degree of bank erosion, the South Morava River is rich with a great amount of material that settles in the riverbed, while runoff from arable land is responsible for the increased concentration of organic matter, pesticides, and fertilizers. These processes change the physical and chemical composition of water. The river flows through an agricultural region in which a large number of villages and farms are located [27-29].

In most industrial centers (Vranje, Vladičin Han, Leskovac, and Niš town), there are no plants for the treatment of industrial and municipal wastewater. Landfills of organic and inorganic waste on its banks are another pollution source of the South Morava River was.

The grouping of monitoring stations by similarity in relation to the parameters defined by the extracted VF factors was obtained by cluster analysis and shown by dendrograms in Figure 2.

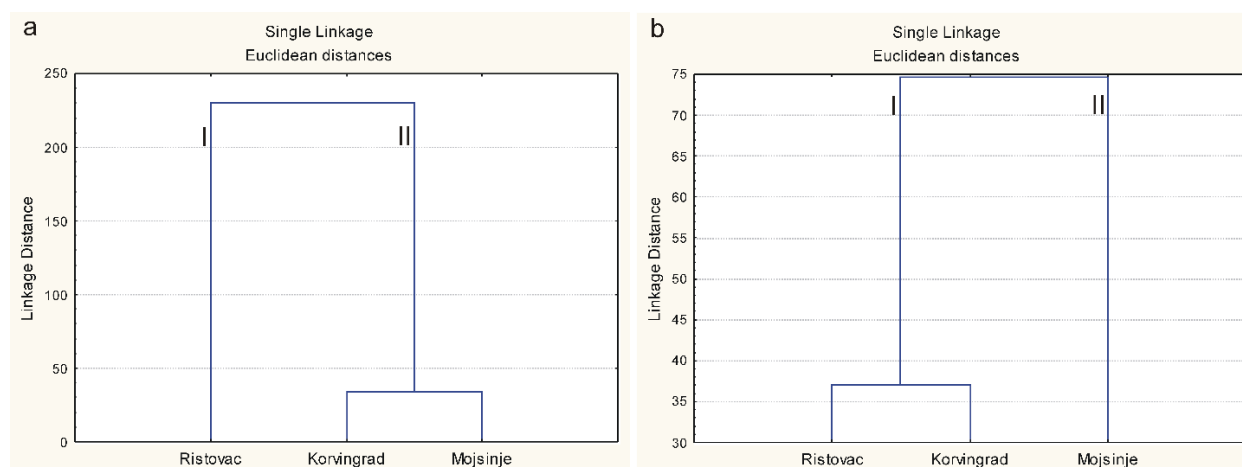


Figure 2. Dendrogram in relation to VF1 (a) and VF2 factor (b)

Slika 2. Dendrogram u odnosu na VF1 (a) i VF2 faktor (b)

Two clusters were noticed in the dendrogram obtained in relation to the VF1 factor by hierarchical cluster analysis. Monitoring station Ristovac is grouped in cluster 1, while monitoring stations Korvingrad and Mojsinje are grouped in cluster 2 (Figure 2a). The cluster analysis indicated that the Ristovac station is more polluted compared to other monitoring stations. Probable pollution sources that deteriorate the water quality on the Ristovac station are the inflow of industrial and municipal wastewater and runoff from the agricultural land. In the paper of Voza et al. [4], the Ristovac station was marked by cluster analysis as a "high polluted" at Morava River system for the investigated period, from 2005 to 2012. The Serbian Water Quality Index (SWQI) method classified the Ristovac station with the worst index number, SWQI=69 ("bad water") for 2009 [25].

Cluster 1 in the dendrogram, which is obtained in relation to the VF2 factor, consists of Ristovac and Korvingrad station, and cluster 2 represents Mojsinje station (Figure 2b). The effects of bank erosion and leaching of the terrain were pronounced at the Mojsinje station. Based on the values of T, SS, TP, BOD₅, and TOC parameters, it was concluded from Table 3 that the process of bank erosion and leaching of the terrain had an influence on pollution in the sector watercourse Ristovac.

The results obtained by the WPI statistical method are given in Table 5. In 2015, the water of the South Morava River was variable in quality, WPI values were in a range from 1.908 to 2.488 (Table 5). The WPI index reflected the combined results of physical and chemical parameters as a single value [22], which was the highest at the Ristovac station, WPI=2.488 - a descriptive indicator of "polluted" water. The exit monitoring profile (Mojsinje) had a lower WPI value, 2.274, and better water quality compared to the entry monitoring profile (Ristovac). The surface water of the South Morava River was less polluted at the Korvingrad station, where water was classified in class III, "moderately polluted". The Danube River WPI index value determined in 2014 for 10 monitoring stations (WPI=1.125-1.539) indicated "moderately polluted" water body, which water quality corresponded to class III [23].

Ecological status includes the quality of the structure and functioning of aquatic ecosystems, related to surface waters, and is classified by Annex V of EU WFD. In Serbia, ecological status is more precisely determined by Regulation on the parameters of ecological and chemical status of surface water and parameters of the chemical and quantitative status of groundwater (Official Gazette of the Republic of Serbia, No. 74/2011). The regulation describes the methodology for determining the ecological status of rivers and

lakes, and the ecological potential for heavily modified and artificial water bodies depending on the defined physical, chemical, and biological parameters. The ecological status/potential of

water at the monitoring stations of the South Morava River was determined with high-level reliability for 9 individual physical and chemical parameters and represented by Table 6.

Table 5. WPI index of water at the monitoring stations of the South Morava River

Tabela 5. WPI indeks vode na monitoring stanicama reke Južne Morave

Parameter		SFQS	Monitoring station		
			Mojsinje	Korvingrad	Ristovac
			$(Ci/SFQS) \times (1/n)$		
pH	-	8.50	0.105	0.106	0.102
DO	mg/L	8.50	0.133	0.143	0.116
NH ₄ -N	mg/L	0.05	0.244	0.222	0.311
NO ₃ -N	mg/L	1.50	0.095	0.090	0.131
PO ₄ -P	mg/L	0.02	0.478	0.439	0.600
TP	mg/L	0.05	0.675	0.442	0.653
BOD ₅	mg/L	1.80	0.208	0.170	0.220
TOC	mg/L	2.00	0.311	0.266	0.320
Cl ⁻	mg/L	50.00	0.025	0.030	0.035
WPI = $\sum (Ci/SFQS) \times (1/n)$			2.274	1.908	2.488

Table 6. Ecological status/potential of water at the monitoring stations of the South Morava River

Tabela 6. Ekološki status/potencijal vode na monitoring stanicama reke Južne Morave

Parameter		Monitoring station		
		Mojsinje	Korvingrad	Ristovac
pH	-	I	II	II
DO	mg/L	I	II	II
NH ₄ -N	mg/L	III	II	III
NO ₃ -N	mg/L	I	II	II
PO ₄ -P	mg/L	II	II	III
TP	mg/L	III	II	III
BOD ₅	mg/L	II	II	II
TOC	mg/L	III	II	III
Cl ⁻	mg/L	I	II	II
Ecological status/potential		III	II	III

In the case of monitoring station Mojsinje, three parameters exceed the limit values for II class, NH₄-N, TP, and TOC, and therefore the water corresponds in quality to class III and has moderate ecological status (Table 6). The regulation was prescribed strictly in article 5 and paragraph 5 the following rule for determining the status of surface waters: "If one or more parameters of ecological status/potential exceed the limits of good status, ecological status/potential of surface waters can be mostly classified as "moderate". Moderate ecological status is characterized by values of quality elements that

deviate moderately from undisturbed status of surface water body type resulting from human activity [21]. The final ecological potential of water at the monitoring station Korvingrad was evaluated as "good" (class II), while at the Ristovac station there was a deterioration of water quality, and the ecological potential was classified in a worse class, III - "moderate potential". At the Ristovac station, 4 physico-chemical parameters, NH₄-N, PO₄-P, TP, and TOC, do not ensure the normal functioning of the river ecosystem as is the case with the water of Korvingrad station. The identified "moderate ecological status/potential" at Mojsinje and Ristovac station provide information on the disturbed natural condition of the South Morava in these sectors of a watercourse, and there is a need for sanction measures and to adhere to certain existing regulations that would enable achieving good watercourse status.

From the presented statistical and comparative analyzes, it was noticed that the watercourse at the Ristovac station was under the greatest load of organic matter and nutrients, and also, there were significant amounts of suspended matter and minerals in the water. Therefore, the further subject of the paper is a complex investigation of the Ristovac station water quality over a longer period. Table 7 shows the average annual values of water quality parameters Ristovac station from 2011 to 2018.

FA/PCA analysis was applied to average annual values of water parameters of the Ristovac station (Table 7), and four VF factors were identified. Factors are responsible for the variability of water quality up to 90.874% (Table 8).

Table 7. Average annual values of water quality parameters at monitoring station Ristovac for period from 2011-2018

Tabela 7. Srednje godišnje vrednosti parametara kvaliteta vode na monitoring stanici Ristovac za period od 2011-2018. godine

Parameter	Unit	Year							
		2011	2012	2013	2014	2015	2016	2017	2018
T	NTU	35.89	134.21	34.56	86.14	69.71	118.39	43.12	54.74
SS	mg/L	33.64	108.45	23.91	106.54	63.92	164.50	106.75	54.33
DO	mg/L	9.09	8.35	8.57	8.09	8.88	8.88	9.53	9.08
pH	-	7.78	7.76	7.87	7.79	7.84	7.85	7.92	7.92
EC	mS/cm	679.09	618.18	636.75	538.73	607.92	578.67	603.83	590.08
HCO ₃ ⁻	mg/L	364.73	339.82	309.42	257.64	305.42	294.83	293.00	292.42
Ca ²⁺	mg/L	80.36	85.31	79.33	68.59	82.08	79.22	78.62	78.65
Mg ²⁺	mg/L	24.00	27.16	22.67	21.56	22.58	17.07	16.82	16.54
Cl ⁻	mg/L	24.80	27.37	23.92	15.19	15.77	12.54	16.48	18.33
SO ₄ ²⁻	mg/L	57.64	61.18	55.75	37.36	46.25	39.50	41.00	43.42
NH ₄ -N	mg/L	0.47	0.14	0.25	0.25	0.14	0.13	0.19	0.15
NO ₃ -N	mg/L	0.85	1.00	1.45	1.49	1.77	1.54	1.76	1.87
PO ₄ -P	mg/L	0.227	0.170	0.280	0.166	0.108	0.096	0.127	0.129
TP	mg/L	0.396	0.614	0.361	0.497	0.294	0.472	0.413	0.415
BOD ₅	mg/L	3.01	3.67	3.84	3.56	3.57	3.68	3.97	4.13
TOC	mg/L	7.60	7.59	5.60	6.89	5.76	5.48	6.51	6.41

Table 8. Loadings of a varimax factor for analysed quality parameters of water at Ristovac station

Tabela 8. Opterećenja varimaks faktora za analizirane parametre kvaliteta vode na monitoring stanici Ristovac

Parameter	VF1	VF2	VF3	VF4
T	-0.087	-0.916	-0.344	-0.023
SS	-0.496	-0.700	-0.210	-0.349
DO	-0.206	0.691	-0.382	-0.525
pH	-0.676	0.640	-0.223	0.127
EC	0.789	0.527	-0.258	-0.137
HCO ₃ ⁻	0.869	0.175	-0.378	-0.248
Ca ²⁺	0.431	0.138	-0.874	-0.018
Mg ²⁺	0.837	-0.326	0.020	0.240
Cl ⁻	0.894	0.043	-0.192	0.320
SO ₄ ²⁻	0.915	0.065	-0.313	0.243
NH ₄ -N	0.662	0.324	0.566	-0.330
NO ₃ -N	-0.906	0.310	-0.072	0.183
PO ₄ -P	0.700	0.280	0.362	0.442
TP	0.168	-0.845	-0.146	0.045
BOD ₅	-0.670	0.129	-0.363	0.522
TOC	0.640	-0.322	0.116	-0.247
Eigenvalue	7.292	3.770	2.091	1.387
%Total variance	45.573	23.562	13.068	8.671
Cumulative % variance	45.573	69.135	82.203	90.874

The first VF1 factor explains 45.573% of the total variance and was correlated (loading>0.50) with pH, EC, HCO₃⁻, Mg²⁺, Cl⁻, SO₄²⁻, NH₄-N, NO₃-N, PO₄-P, BOD₅, and TOC. The factor is represented by the influence of natural sources of pollution (a mineral component of river water - the dissolution of constituents of the South Morava riverbed) and anthropogenic diffuse (runoff from the agricultural land), and point sources of pollution (industrial and communal wastewater) [9,42].

The VF2 factor contributed 23.562% in total variance. It is characterized by a strong negative loading for T and TP, moderate negative loading for SS, and it has moderate positive loads on DO, PH and EC. This factor is an obvious example of the impact of bank erosion and terrain leaching on the quality of surface water.

The VF3 factor accounted for 13.068% of the total variance and was strong negatively associated with Ca²⁺ and moderate positively with NH₄-N. Higher concentrations of ammonia in surface water originate from municipal and industrial wastewater [42], but the effluents also contain anions that interact with potassium and reduce its concentration in water.

The VF4 factor, which explains 8.671% of total variance, has moderate positive loading on BOD₅ and moderate negative loading on DO. During the degradation process of organic matter by microorganisms, the oxygen concentration decreases [40], so that the factor can be described as a load of watercourses with biodegradable organic pollution.

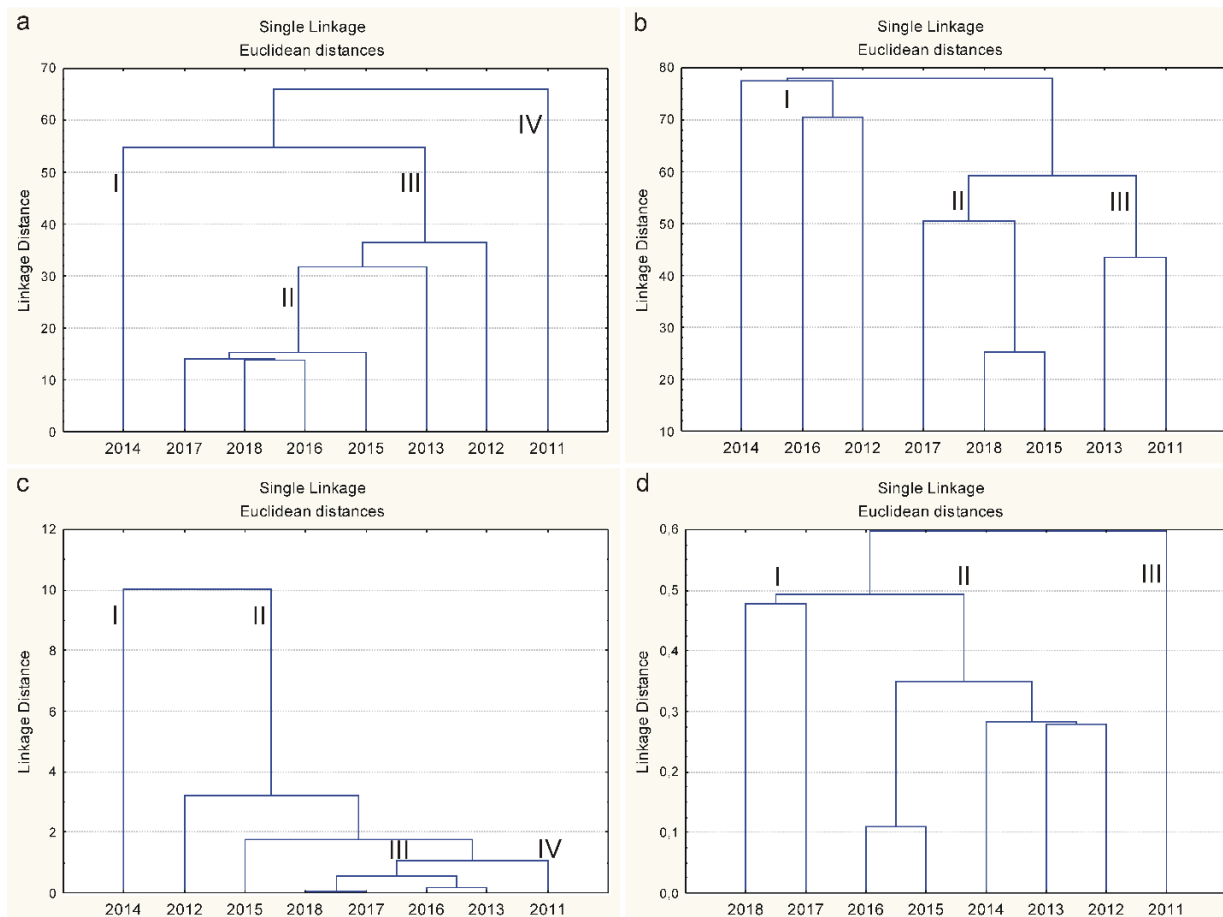


Figure 3. Dendrograms in relation to VF1 (a), VF2 (b), VF3 (c) and VF4 (d) factor

Slika 3. Dendrogram u odnosu na VF1 (a), VF2 (b), VF3 (c) i VF4 (d) factor

Cluster analysis in relation to VF1 factor generated four groups of investigated years by similarities: cluster I - 2014, cluster II - 2015, 2016, 2017 and 2018, cluster III - 2012 and 2013, and cluster IV - 2011 (Figure 3a). In the area in front of the Ristovac station, there are two settlements Preševo and Bujanovac, which are some of the main polluters of the South Morava River. Runoff of wastewater from the settlement to the recipient, the South Morava River, is in most cases without prior treatment, and besides, the result of the population activity is the appearance of a larger number of illegal landfills, which "sprout" along the river bank. In 2011, a significant effect of mineralization of geological components of the soil due to the exchange of soil and water cations was recorded (cluster IV). The quality of the watercourse at the Ristovac station in 2011 was also determined by the inflow of wastewater, although the impact of runoff from agricultural land could not be excluded. Poor water quality was observed in 2012 and 2013 (cluster III) as a function of mineral components, $\text{NH}_4\text{-N}$, $\text{PO}_4\text{-P}$ and TOC (Table 7). Surface water in 2014 is characterized by the lowest mineral content (cluster I).

The hierarchical cluster analysis performed in relation to the VF2 factor, groups the years of water quality testing at the Ristovac station into 3 clusters: 2012, 2014, and 2016 (cluster I), 2015, 2017 and 2018 (cluster II) and 2011 and 2013 (cluster III), Figure 3b. In the upper part of the South Morava basin, from the settlement of Grdelica towards the border with Northern Macedonia, erosion of medium intensity occurs. Also, in this area, illegal exploitation of sand and gravel from the South Morava riverbed is carried out [26]. In 2012, 2014, and 2016 (cluster I) compared to other examined years, agricultural soil water erosion was more expressed, which contributes to a significant increase of TP concentration in the water body (Table 7). At the Ristovac station, the lowest degree of bank erosion and terrain leaching were identified in 2011 and 2013 (cluster III, Table 7).

From data sets, in relation to the VF3 factor, four groups of examined years were obtained on a dendrogram: cluster I - 2014, cluster II - 2012 and 2015, cluster III - 2013, 2016, 2017, 2018, and cluster IV - 2011 (Figure 3c). Once again, the

impact of point source pollution, industrial or municipal effluents, on water quality at the monitoring station Ristovac was observed in 2011 (cluster IV). In this year, the highest water load by ammonia was recorded, 0.47 mg/L (Table 7). The 2012 and 2015 years are grouped in cluster II, which is characterized by lower effects of wastewater, i.e. low $\text{NH}_4\text{-N}$ content (0.14 mg/L), and highest Ca^{2+} content (82.08 and 85.31 mg/L). The examined years included in cluster III were grouped primarily according to the uniform content of the mineral component ($\text{Ca}^{2+}=78.62\text{-}79.33$ mg/L). The lowest content of Ca^{2+} ions, 68.59 mg/L, exists in the water of South Morava in 2014 (cluster I).

In relation to the VF4 factor, cluster analysis shows a dendrogram with three clusters: I - 2017 and 2018, II - 2012, 2013, 2014, 2015 and 2016, and III - 2011 (Figure 3d). The highest amount of

biodegradable organic waste existed in the water at the Ristovac station in 2017 and 2018 (cluster I), while the lowest concentration of biodegradable matter was recorded in 2011 (cluster III).

Grouping of the investigated years based on VF factors obtained by FA/PCA analyzes helped to extract and identify the factors responsible for changing water quality in the observed period. In 2011, water quality was under the greatest influence of natural and anthropogenic pollution factors. The effect of bank erosion was the dominant process in 2012, 2014, and 2016 at the Ristovac station. In addition to 2011, the 2012 and 2013 showed poor water quality, while in 2015 the surface water was the purest.

The degree of surface water pollution through WPI value was represented by Table 9.

Table 9. WPI index of water at the monitoring station Ristovac

Tabela 9. WPI indeks vode na monitoring stanici Ristovac

Parameter	Year							
	2011	2012	2013	2014	2015	2016	2017	2018
	(Ci/SFQS) × (1/n)							
pH	0.102	0.101	0.103	0.102	0.102	0.103	0.103	0.103
DO	0.119	0.109	0.112	0.106	0.116	0.116	0.125	0.119
$\text{NH}_4\text{-N}$	1.044	0.311	0.555	0.555	0.311	0.289	0.422	0.333
$\text{NO}_3\text{-N}$	0.063	0.074	0.107	0.110	0.131	0.114	0.130	0.138
$\text{PO}_4\text{-P}$	1.261	0.944	1.555	0.922	0.600	0.533	0.705	0.717
TP	0.880	1.364	0.802	1.104	0.653	1.049	0.918	0.922
BOD_5	0.186	0.226	0.237	0.220	0.220	0.227	0.245	0.255
TOC	0.422	0.422	0.311	0.383	0.320	0.304	0.362	0.356
Cl^-	0.055	0.061	0.053	0.034	0.035	0.028	0.037	0.041
WPI	4.132	3.612	3.835	3.536	2.488	2.763	3.047	2.984

WPI value calculated based on physical and chemical parameters for 2011 corresponds to class V, "impure" surface water. The water at the Ristovac station from 2012-2018 had the character of "polluted" water (WPI=2.488-3.835), class IV. The water body South Morava in 2015 suffered the least pollution (WPI=2.488) in terms of analyzed

parameters. A tendency water quality improvement occurred at the Ristovac station from 2011 to 2018 (Table 9). Based on the value of the WPI index, the two largest rivers in Serbia (the Danube and Sava River) were characterized as "moderately polluted" and "polluted" surface water (class III and IV) [22,23].

Table 10. Ecological potential of water at the monitoring station Ristovac

Tabela 10. Ekološki potencijal vode na monitoring stanici Ristovac

Parameter	Year							
	2011	2012	2013	2014	2015	2016	2017	2018
pH	II	II	II	II	II	II	II	II
DO	II	II	II	II	II	II	II	II
$\text{NH}_4\text{-N}$	III	III	III	III	III	III	III	III
$\text{NO}_3\text{-N}$	II	II	II	II	II	II	II	II
$\text{PO}_4\text{-P}$	IV	III	IV	III	III	II	III	III
TP	III	IV	III	IV	III	IV	IV	IV
BOD_5	II	II	II	II	II	II	II	II
TOC	IV	IV	III	III	III	III	III	III
Cl^-	II	II	II	II	II	II	II	II
Ecological potential	IV	IV	IV	IV	III	IV	IV	IV

Relevant parameters for determination of ecological quality (potential) were used in classification water of the South Morava River at the monitoring station Ristovac by Regulation on the parameters of ecological and chemical status of surface water and parameters of the chemical and quantitative status of groundwater (Official Gazette of the Republic of Serbia, No. 74/2011). Obtained results of the ecological potential of water at Ristovac station (Table 10) confirmed WPI statistical assessment of watercourse pollution.

The altered natural surface water body, the South Morava River, in the investigated region, is generally classified in class IV, "poor ecological potential", which significantly deviated from the required good potential, class II. Only water in 2015 was in the scope of classes II and III. The parameters that were partly responsible for surface water poorer quality by regulation in all investigated years, except 2015, were: $\text{PO}_4\text{-P}$, TP, and TOC. This monitoring analysis predicts temporal variations of water quality, which will serve to set a more rational watercourse management strategy in the following years.

4. CONCLUSIONS

In this paper, the combined approach by different multivariate statistical methods and quality indicators (ecological status/potential and WPI) enabled the assessment of the South Morava River pollution and the identification of pollution sources.

FA/PCA method interprets and represents data in adequate two-factor (monitoring stations) or four-factor model (years), which describes variation in the water quality of the South Morava River, while CA method helps to determine key polluted stations and years based on similarity. The physical and chemical composition of the water at the monitoring stations (Mojsinje, Korvingrad, and Ristovac) along the South Morava River were most influenced in 2015 by industrial and sewage effluents, runoff from agricultural land, and the process of erosion. The dominant effects which deteriorate the water quality at Mojsinje station were bank erosion and leaching of the terrain, but at Ristovac station those were an inflow of wastewater without prior treatment, runoff from arable land, and the dissolution of constituents of the South Morava riverbed. The high pollution level of water at Ristovac station indicated the need for eight-years monitoring, 2011-2018. Multivariate analysis showed that water in 2011 suffered from a high load of nutrients ($\text{NH}_4\text{-N}$ and $\text{PO}_4\text{-P}$) and organic matter. In 2012, 2014, and 2016, the effect of erosion and leaching of the terrain was represented.

The EU WFD established a base for monitoring surface waters (rivers, lakes), as well as the

possibility of adapting monitoring programs in Serbia and other countries, which are reflected in the assessment of the ecological status/potential of water bodies. The determined ecological status of the South Morava water had differences between stations, Korvingrad achieved a "good ecological potential", but Mojsinje and Ristovac exceeded the limits of the required status water body by EU WFD (moderate ecological status/potential). The summary ecological status of water at Ristovac station in the observed period 2011-2018, except 2015, generally corresponds to "poor ecological potential". The parameters contributing to failing surface water to achieve a "good ecological potential" are $\text{PO}_4\text{-P}$, TP, and TOC.

The assessment of surface water quality was completed by the index method (WPI), which classified water at Mojsinje and Ristovac station as "polluted" (class IV) and at Korvingrad as "moderately polluted" (class III). WPI values have shown a high pollution level of the South Morava River at Ristovac station (class IV-V), and pollution degree was generally decreased from 2011-2018. The 2011 year was marked as the most polluted, class V ("impure" surface water).

FA/PCA and WPI methods represent a useful tool in reliable identification and classification of pollution in surface water at the analyzed object (stations and years). Ecological status obtained by prescribed regulative and EU WFD effectively determines the deviation from the required standard, which indicates the application of an appropriate monitoring strategy and sanction measures. The solution for the protection of the water quality of the South Morava River, primarily at Ristovac station, in general lies in the regulation system of communal and industrial wastewater, construction of adequate wastewater treatment plants, conducting anti-erosion measures, and prevention of formation of illegal landfills on banks.

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IZVOD

ODREĐIVANJE ZAGAĐENJA VODE REKE JUŽNE MORAVE (SRBIJA) STATISTIČKIM I INDEKSNIM METODAMA

Faktorska analiza/analiza glavnih komponenti (FA/PCA), primenjena na 16 fizičko-hemijskih parametara sa tri različita monitoring mesta (Mojsinje, Korvingrad i Ristovac) reke Južne Morave u 2015. godini i za period od 2011-2018. godine za stanicu Ristovac, je izdvojila dva odnosno četiri varimaks faktora koji objašnjavaju 100,000 i 90,874% ukupne varijanse kvaliteta vode, redom. Veza između stanica i godina dobijena klaster analizom (CA) kategoriše posmatrane objekte prema različitim nivoima kvaliteta. Parametri odgovorni za zagađenje reke Južne Morave odnose se na tačkasti (industrijski/kanalizacioni efluenti), netačkasti (oticanje sa obradivog zemljišta i erozija) i prirodni izvor (mineralna komponenta rečne vode). Rezultati metode indeksa zagađenja vode (WPI) za 2015. godinu pokazali su da reka Južna Morava na stanici Korvingrad predstavlja „umereno zagađeno“ vodno telo (klasa III), dok na stanici Mojsinje i Ristovac „zagađeno“ vodno telo (klasa IV). Dobijene vrednosti ekološkog potencijala ukazuju da kvalitet vode na stanici Korvingrad odgovara klasi II, „dobar ekološki potencijal“. Površinska voda na stanicama Mojsinje i Ristovac odstupa od propisanih ekoloških standarda Okvirne direktive o vodama Evropske unije (EU WFD), klasa III. Reka Južna Morava ima najveće opterećenje nutrijentima (PO₄-P i TP) i organskom materijom (TOC) na stanici Ristovac, što je potvrđeno indeksom i uporednom metodom (zagađena voda sa slabim ekološkim potencijalom) u periodu od 2011-2018. godine. Sveobuhvatnom analizom rada utvrđen je trend smanjenja zagađenja reke Južne Morave, pre svega na stanici Ristovac.

Ključne reči: reka Južna Morava, kvalitet vode, multivarijacione statističke metode, indeks zagađenja vode, ekološki status/potencijal

Naučni rad

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