Desalination and Water Treatment
www.deswater.com
doi: 10.5004/dwt.2023.29307

285 (2023) 67–77
February

Occurrences of water quality assessment using improvised water quality index at the Danube River, Serbia

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Received 12 July 2022; Accepted 18 January 2023

ABSTRACT

The water quality index is very often used to classify the water quality of a river. The paper aims to develop a new improvised water quality indicator using statistical methods. To reduce the number of analysed chemical parameters and simplify the tests, WQI_{min} was used. The results of the research conducted in Serbia in 2019 were subjected to correlation and principal component analysis (PCA). This reduced the number of parameters from 29 to 8, significantly reducing the time and cost of laboratory analyses. The temporal and spatial variability evaluation can be performed using WQI_{min}, which consists of dissolved oxygen, total phosphorus, orthophosphate, total dissolved solids, suspended solids, chlorides, ammonium, and nitrite nitrogen. The main sources of river contamination were untreated industrial sewage, fish farming wastewater, and soil erosion. The highest level of pollution was observed in winter. Good water quality was found in the river's upper course and very good in the lower course. Water quality values obtained based on a complete and simplified set of parameters were strongly correlated with each other.

Keywords: Danube River; Serbia; WQI; Water quality assessment

1. Introduction

There are about 1.39 billion km^3 of water on earth, of which only 0.12% is freshwater used for human supply and industrial and agricultural production. According to the World Health Organisation, about eight million people worldwide cannot access to clean drinking water. As emphasized by the World Bank, 2 billion people worldwide live in countries with a drinking water shortage, and this number may double in the next two decades [1–3]. These problems found their place in international documents [4–7]. Many natural and anthropogenic factors affect the availability of water. The main causes of environmental pollution are primarily:

- intensive farming with the use of fertilizers and pesticides [8–11];
- industry generating dust and wastewater [12,13];
- transport contributing to exhaust gas emissions [14,15];
- the use of conventional energy sources for heating households [16,17];
- housing estates without sewage systems or sewage treatment plants [18–20];
- poorly secured landfills [21,22].

In Serbia, surface waters are used for various ecosystem services, for example, drinking, hydropower, irrigation, transport, and fishing. The Danube is the most important source of drinking water for around ten million people and

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is the primary source of drinking water, supplying cities in Germany and Romania. The water supply for the population and industry in Serbia is currently distributed by many local groundwater water intakes. Approximately 30% of electricity is generated by hydropower plants, the largest of which can be found on the Danube River. Irrigation in agriculture has an extensive but insufficiently exploited potential. The soil and climatic conditions favour cereals, industrial and forage crops, fruit, and vegetables. The same is valid for waterborne transport, accounting for up to 5% of total freight transport in Serbia. However, passenger traffic and the number of cruise ships have grown over the past two decades. Fish farming is also a promising ecosystem service in Serbia, which, together with fishing in surface waters, covers almost half of the national needs [23–29].

The analysis of water quality indicators intended for supplying the population indicates a systematic improvement in drinking water quality standards. They change depending on the current state of knowledge and the local economic and social conditions. On the one hand, this is due to the presence of a decreasing number of pollutants in the abstracted waters and better analytical possibilities. On the other hand, attention is paid to determining the impact of substances on the health of the people and the importance of protecting water resources. WHO guidelines are recognised worldwide as the most reliable information on standard drinking water quality and form the basis for national regulations and laws [30–33].

Surface and groundwater quality is fundamental to economic and civilization development and indispensable in preserving natural values. Sometimes it is used conventional methods, which consist of comparing the determined parameter values with the existing pollution standards. However, we often deal with a large data set when interpreting individual parameters is difficult. Then, the water quality index (WQI) is used to manage water resources because we get a single number that allows us to simplify a complex set of river water quality parameters. The first in the world, methodologically presented in the literature, was the water quality index developed by Horton. The development of knowledge in the following years led to the formulation of various WQI variants. The most commonly used are the Weighted Arithmetic Water Quality Index (WQI), the US National Sanitation Foundation Water Quality Index (NSFWQI), the Canadian Council of Ministers of the Environment Water Quality Index (CCMEWQI), the British Columbia Water Quality Index (BCWQI), and Oregon Water Quality Index (OWQI) [34–36].

The water quality assessment in 2019 at nine locations was carried out due to the great economic importance of the Danube. Surface water provides many ecosystem services serving people: hydropower, shipping, fish farming, irrigation of crops, and freshwater supplies. The article aims to analyse the river's water quality and develop a water quality index, including a minimum data set. In this study, the Weighted Arithmetic Water Quality Index (WQI) will be used to assess the water quality in the Danube River in Serbia. The procedure for developing a new modified index will be carried out in six stages. The first step involves selecting 29 water quality parameters and creating a database for them. The second stage includes statistical processing of the results from the created database. The third step involves computing the WQI for the complete data set. The fourth step is to establish the main components of WQI_{full}. The fifth step is to establish a minimum set of parameters and calculate WQI_{min}. The sixth step is to compare WQI_{full} and WQI_{min}.

2. Material and methods

2.1. Study area

The Danube is the second longest river in Europe. The river flows west to east through many European geographic regions, including the Bavarian Highlands, the Pannonian Basin, and the Wallachian Lowlands. Its basin covers large parts of Central and South-Eastern Europe. The river head is located in the Black Forest mountains. The length river amounts to 2,826 km, the catchment area is 817,000 km^2, and the average flow is 6,700 $m^3 \cdot s^{-1}$. The Danube has a flow path of ten European countries: Germany, Austria, Slovakia, Hungary, Croatia, Serbia, Bulgaria, Romania, Moldova, and Ukraine, to mouth the Black Sea. The river length in Serbia is 587 km. Like other rivers, the Danube has met anthropogenic influences since the beginning. Only 20% of the 19th-century floodplains have survived to the present day, and only 25% of the river's course can now be described as close to nature. The intensive development of agriculture, industry and tourism contributed to the increase in water pollution in the 20th century. This resulted in the discharge of large amounts of untreated sewage. The capitals of three countries (Belgrade, Budapest, and Wien) are found on the river. In addition, biological life in the river was destroyed by hydrotechnical structures (Iron Gate on the border of Serbia and Romania). International protection is difficult to implement, as all countries want to use the river economically and reap the benefits of its geographical location.

The Middle Danube Region is mainly characterised by the continental climate. Serbia is situated on the border of two climatic zones, warm temperate in the north and subtropical in the south. The terrain is highly geographically differentiated. Most of the southern rim of the Pannonian Lowlands is arable land and is relatively uniform and plain (80%). The Pannonian Basin is bounded to the west by the Alps, to the north and east by the Carpathians, and to the south by the Dinaric Mountains. It is a tectonic sinkhole between the Alps and the Carpathians. Agriculture is the source of wealth in these areas. Fertile soils and a mild climate make the Pannonian Basin an agricultural and breeding area since ancient times. The main river tributaries on this part are the Drava, Sava, Tisa, and Velika Morava. Winter temperatures range from −1°C to +3°C, and frosts are frequent. In summer, it is hot, with average values ranging from 23°C to 28°C. In the lowlands, there is an average rainfall of 600–700 mm, while in the mountains, the rainfall is higher and amounts to 1,000 mm. In the Pannonian Lowlands, there are Chernozems, and in the river valleys are Histosols [37–39].

2.2. Water sampling

Water quality analyses were performed at nine stations on the Danube River in 2019, with a monthly frequency – of 12 samples at each station (Table 1, Fig. 1). Three

Table 1
Location of monitoring stations in Serbia

Station	Number	Mouth	a.s.l.	Longitude N	Latitude E
Bezdan	S1	1,425	79	49°50′33.1″	18°55′13.6″
Bogojevo	S2	1,367	78	45°31′19.5″	19°05′18.1″
Novi Sad	S3	1,255	77	45°13′03.4″	19°48′54.7″
Zemun	S4	1,147	75	44°49′42.8″	20°27′57.4″
Smederevo	S5	1,116	65	44°41′33.3″	20°57′18.7″
Banatska Palanka	S6	1,077	58	44°49′17.7″	21°20′51.6″
Tekija	S7	956	52	44°40′57.5″	22°24′08.5″
Brza Palanka	S8	884	37	44°28′40.8″	22°40′48.1″
Radujevac	S9	852	34	44°16′24.5″	20°27′57.4″

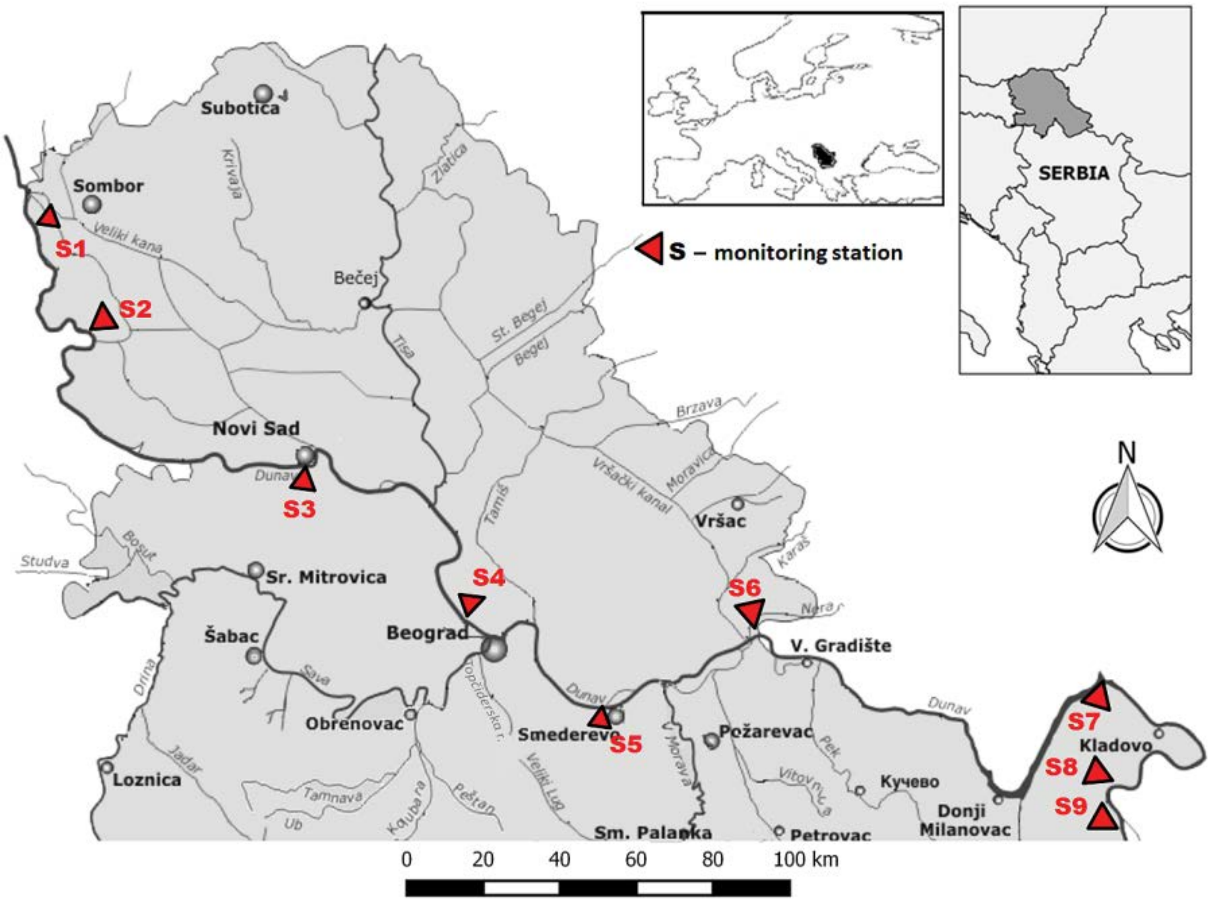

Fig. 1. Locations of monitoring stations along the Danube in Serbia.

samples were collected at each station at different depths. In each sample, 29 water quality parameters were analyses. Chemical analyses were carried out using a PC spectrophotometer (AQUALYTIC, Langen, Germany). The dissolved oxygen (DO), water temperature (WT), electrical conductivity (EC), and pH were measured at the site using a portable multimeter. In the laboratory, nutrients: ammonium nitrogen (NH_4–N), nitrite nitrogen (NO_2–N), nitrate nitrogen (NO_3–N), total nitrogen (TN), orthophosphate (PO_4–P), total phosphate (TP), and desolation: [chloride (Cl^-), sulphates (SO_4^{++}), were measured spectrophotometrically [40]. Suspended solid (SS) and total dissolved solids (TDS) were analyses by gravimetric method, total organic carbon (TOC), using a TOC1200 analyzer. The total hardness (TH) was determined by the edetate method. In filtered metals: (Na^+, Ca^{++}, Mg^{++}, K^+, Fe^{+++}, Mn^{++}), heavy metals: (Zn^{++}, Cu^{++}, Pb^{++}, Cd^{++}, Ni^{++}, Cr^{++}, Al^{++})

were measured by the AAS method [41]. Only the total phosphorus and nitrogen were performed the following prior oxidation of the investigated sample in a thermoreactor at a temperature of 120°C.

2.3. Water quality index

The basis for assessing surface and groundwater quality can be the analysis of the size of selected physical, chemical, and biological properties. However, we use the water quality index much more frequently to manage our water resources. WQI is one value that describes water quality taking into account its temporal and spatial variability. Based on the WQI value, we can decide to use the water for ecosystem services, for example, supplying people with drinking water, recreation, and fish farming. We applied the calculation method using the Weighted Arithmetic Water Quality Index Method in the article. The indicator is quite flexible, allowing the analysed water quality parameters to be selected. It presents the results of the water quality assessment based on its suitability for consumption. WWQI is calculated based on sub-indices as a function of the measured concentration and the concentration standard value. The relevant concentration limits are based on Directive 98/83/EC and the World Health Organization [31,42]. The WQI ranges from 0 to 100 and grades the water quality into five classes: excellent (91–100), good (71–90), medium (51–70), bad (26–50), and very bad (0–25) [43,44]. For the determination of the WQI, the empirical equation was used:

$$\text{WQI} = \frac{\sum C_i \times P_i}{\sum P_i} \tag{1}$$

where C_i is the value assigned to parameter i after normalization, and P_i is the relative weight of parameter i.

2.4. Statistical analysis

All technical conditions for quantifying chemical compounds are met by the applicable standards [30,40]. The results obtained from the research have been interpreted using various statistical methods. One-way ANOVA, Pearson's correlation analysis, and principal component analysis were performed in this study. The ANOVA analysis was indicated for each parameter separately (the hypothesis that the mean values of the examined parameters did not differ significantly was verified), with statistical differences between stations and seasons at $p < 0.05$ (Duncan's test). Using the Shapiro–Wilk test, the consistency of the distribution of the tested water parameters with the normal distribution was examined. To simplify WQI calculations and reduce the number of parameters and costs of analytical detection, principal components analysis (PCA) and correlation analyses were performed. The aim of the PCA and correlation analysis was to reduce the data space's size and identify the water quality parameters that are most responsible for the variability of the tested data set. The original data were standardized to avoid misclassification associated with different units. The results of these analyses were used in the modification of the WQI. Statistical analysis was performed for 108 samples, in which a total of 3024 determinations were performed. The calculations were made using Microsoft Excel and Statistica 13.3.

3. Result and discussion

3.1. Spatial and temporal for seasons variability

The Danube River is often used to supply fish with water, and for this reason, the average concentration of water quality parameters has been compared with the mandatory values specified for inland waters for salmonids and cyprinids [45]. The average concentrations of nutrients determined in all locations did not exceed the mandatory values established for salmon and cyprinids. Only in the case of the SS at site S6, the applicable requirements were exceeded, which should be met by inland waters being fish's living environment in natural conditions. The mean values over the four seasons were calculated for the 29 recorded monthly values of each parameter. In this study, all water quality parameters showed significant seasonal differences (Table 2). Seasonality of changes in temperature and water flow throughout the year cause seasonal changes in water quality, but it was found that four variables (TP, DO, NO_3–N, NH_4–N) are not significantly correlated with the season of the year, rather indicating an anthropogenic impact [46–48].

Table 3 shows the means and standard deviations of the values of 29 water quality parameters recorded in 9 sampling stations. In this study, all variables except pH and EC showed significant spatial changes, indicating the influence of anthropogenic activity [49,50]. For example, the highest NO_3–N, TN, and TP concentrations were found at the S1 station. Also, the S1 station had average concentrations of SO_4^{--}, Fe^{+++}, Mn^{++}, Zn^{++}, Cu^{++}, Ni^{++}, and TOC (Table 3). The sampling station S1 is located near the village of Bezdan, which is situated on the border with Croatia and Hungary. The chemical composition of the pollutants suggests that they come from both agriculture and industry. This is due to the fragmented agriculture farms, the use of artificial fertilizers in the Upper Danube basin countries, and the inadequate functioning of the sewage treatment plant. The cause of the river pollution in this area is the inflow of untreated municipal and industrial wastewater from the city of Pécs (Hungary). There are closed coal and uranium mines nearby, which may emit pollutants in an uncontrolled manner. Another cause of water pollution can be the high concentrations of heavy metals in river sediments in Dunafoldvar town [51]. Significant parts along both banks represent alluvial zones, that is, regularly flooded forests, and meadows. As a result of the processes of self-cleaning and dilution, the concentrations of these parameters decreased with the river current to the S5 station, which improved the water quality [52]. However, the most significant water contamination was found at station S6, where the highest concentrations of SS, TDS, Fe^{+++}, Mn^{++}, Zn^{++}, Cr^{++}, Pb^{++}, and Al^{++} were found (Table 3). The sharp increase in pollution between stations S5 and S6 could result from discharging insufficiently treated wastewater from the metallurgical industry and leaching pollutants from the metal ore mines deposits located below the city of Smederevo. Since the steel plant in Smederevo did not use efficient wastewater treatment processes, the wastewater was characterized by high suspended solids concentrations [53–55]. For this reason,

Table 2
Seasonal variation in water quality in the Danube River in Serbia

Parameter	Winter	Spring	Summer	Autumn
WT (°C)	5.6 ± 2.8^{d}	17.4 ± 5.0^{b}	23.8 ± 1.7^{a}	12.1 ± 4.7^{c}
SS (mg·dm^{-3})	16.7 ± 10.7^{a}	17.8 ± 9.6^{a}	15.4 ± 10.0^{b}	11.4 ± 6.9^{c}
DO (mg·dm^{-3})	11.7 ± 1.1^{a}	8.6 ± 2.1^{c}	7.9 ± 1.2^{c}	9.7 ± 1.1^{b}
TH (mg·dm^{-3})	255.5 ± 79.1^{b}	297.4 ± 96.4^{a}	261.3 ± 103.5^{b}	244.2 ± 86.1^{c}
pH	8.1 ± 0.2	7.9 ± 0.5	8.0 ± 0.3	8.0 ± 0.2
EC (μS·cm^{-1})	442.7 ± 46.3^{a}	352.2 ± 64.9^{c}	363.9 ± 27.2^{c}	419.6 ± 24.6^{b}
TDS (mg·dm^{-3})	259.3 ± 28.3^{a}	218.1 ± 41.4^{b}	216.8 ± 14.6^{b}	248.3 ± 19.2^{a}
NH_4–N (mg·dm^{-3})	0.12 ± 0.06	0.11 ± 0.05	0.09 ± 0.05	0.12 ± 0.05
NO_2–N (mg·dm^{-3})	0.02 ± 0.01	0.02 ± 0.01	0.02 ± 0.01	0.02 ± 0.01
NO_3–N (mg·dm^{-3})	1.60 ± 0.7^{a}	0.91 ± 0.24^{b}	0.78 ± 0.31^{c}	0.97 ± 0.30^{b}
TN (mg·dm^{-3})	3.20 ± 1.0^{a}	1.57 ± 0.54^{c}	1.75 ± 0.86^{b}	1.92 ± 0.76^{b}
PO_4–P (mg·dm^{-3})	0.04 ± 0.01	0.04 ± 0.02	0.05 ± 0.02	0.06 ± 0.02
TP (mg·dm^{-3})	0.10 ± 0.04	0.10 ± 0.05	0.11 ± 0.05	0.12 ± 0.05
Na^+ (mg·dm^{-3})	16.7 ± 4.1^{a}	11.4 ± 3.1^{c}	13.5 ± 3.3^{b}	14.7 ± 2.8^{b}
K^+ (mg·dm^{-3})	2.04 ± 0.75^{b}	2.16 ± 0.78^{b}	2.45 ± 1.14^{a}	2.57 ± 0.69^{a}
Ca^{++} (mg·dm^{-3})	61.1 ± 4.4^{a}	53.3 ± 12.0^{b}	50.9 ± 5.3^{b}	58.5 ± 7.7^{a}
Mg^{++} (mg·dm^{-3})	15.4 ± 3.6^{a}	13.4 ± 4.2^{b}	11.5 ± 2.2^{c}	13.9 ± 3.5^{b}
Cl^- (mg·dm^{-3})	25.7 ± 5.2^{a}	16.2 ± 3.3^{d}	19.2 ± 3.0^{c}	22.9 ± 4.2^{b}
SO_4^{--} (mg·dm^{-3})	32.5 ± 6.8^{a}	28.4 ± 7.1^{b}	28.2 ± 5.4^{b}	30.8 ± 7.9^{a}
Fe^{+++} (μg·dm^{-3})	482.9 ± 260.5^{a}	443.6 ± 171.6^{b}	407.7 ± 169.8^{b}	347.4 ± 118.2^{c}
Mn^{++} (μg·dm^{-3})	35.1 ± 16.2^{b}	45.1 ± 21.4^{a}	31.3 ± 10.2^{b}	23.0 ± 8.2^{c}
Zn^{++} (μg·dm^{-3}	66.5 ± 15.4^{a}	56.4 ± 21.0^{a}	23.2 ± 16.8^{b}	26.1 ± 19.8^{b}
Cu^{++} (μg·dm^{-3})	14.1 ± 6.5^{a}	10.9 ± 6.8^{b}	7.7 ± 3.7^{c}	9.6 ± 7.4^{b}
Cr^{++} (μg·dm^{-3})	1.73 ± 0.68^{b}	2.24 ± 1.00^{a}	1.43 ± 0.89^{b}	0.83 ± 0.49^{c}
Pb^{++} (μg·dm^{-3})	1.31 ± 0.75^{b}	1.44 ± 0.58^{a}	1.02 ± 0.47^{c}	1.49 ± 1.00^{a}
Cd^{++} (μg·dm^{-3})	0.03 ± 0.01	0.08 ± 0.04	0.03 ± 0.01	0.03 ± 0.01
Ni^{++} (μg·dm^{-3})	5.63 ± 2.44^{a}	4.14 ± 2.22^{c}	3.91 ± 3.87^{c}	4.83 ± 2.92^{b}
Al^{++} (μg·dm^{-3})	326.5 ± 115.4^{a}	287.3 ± 90.9^{b}	250.4 ± 116.6^{b}	169.3 ± 82.1^{c}
TOC (mg·dm^{-3})	4.22 ± 0.88^{a}	4.19 ± 1.22^{a}	3.45 ± 0.95^{b}	4.26 ± 1.69^{a}

Various letters in the poem indicate statistical differences between the seasons.

industrial wastewater increases the concentration of SS, Fe^{+++}, and Al^{++} in the water. Another factor that might contribute to the pollution is soil erosion during periods of high flows and the fact that the S6 is located at the mouth of the Danube–Tisa–Danube canal and the Danube. Namely, this huge amelioration canal drains water from adjusted arable land and receives agricultural runoff after heavy rainfall. All of the mentioned factors contribute to the high levels of SS, Fe^{+++}, and Al^{++} in the river [56].

3.2. Water quality assessment using WQI_{full}

Twenty-nine standardized water quality parameters were used to calculate WQI_{full} in nine sites and four seasons in the Danube River. The mean annual WQI value for the river was 90 and ranged from 83.6 to 96. The WQI analysis allowed us to classify the surface water in the river as excellent at stations S3, S4, S5, S7, S8, and S9; as good at stations S1, S2, and S6.

The highest mean annual value of WQI was found at station S5, and it was 96. The lowest mean value of WQI was found at S1 – 83.6. The lowest water quality at S1 results from the inflow of pollutants from Hungary and Croatia. Water quality management in transboundary catchments is a complex and difficult-to-solve economic problem [57]. The sampling point in the middle course of the river (S6) showed a statistically significant ($p < 0.05$) deterioration of water quality by 9 points compared to the higher station (S5). The decline in the suitability of drinking water is related to industrial activity and a greater discharge of poorly treated wastewater below station 5. The increase in nutrient pollution is often caused by the discharge of untreated wastewater and other anthropogenic activities, as confirmed by the results of the Klang River catchment area [58]. Based on our research, we conclude that the water quality in the Danube River was subject to seasonal changes and requires improvement in the upper section. Due to the apparent impact of point pollution, local governments should make more significant efforts to control pollution. Station 9 did not show any differences between the seasons of the year. This may be because of its location below the Iron Gate and its less anthropogenic influence. Relatively larger seasonal fluctuations were observed

Table 3
Spatial variation in water quality in the Danube River in Serbia

Parameter	Bezdan	Bogojevo	Novi Sad	Zemun	Smederevo	Banatska Palanka	Tekija	Brza Palanka	Radujevac
WT (°C)	13.7 ± 7.7^{a}	13.9 ± 8.3^{a}	14.7 ± 8.2^{b}	13.8 ± 7.6^{a}	14.8 ± 7.8^{b}	14.7 ± 8.6^{b}	15.6 ± 8.2^{c}	15.7 ± 8.2^{c}	16.1 ± 6.9^{c}
SS ($mg \cdot dm^{-3}$)	24.08 ± 14.9^{c}	22.91 ± 12.3^{c}	23.50 ± 12.55^{c}	15.11 ± 6.11^{b}	10.82 ± 6.59^{b}	28.92 ± 16.3^{c}	5.50 ± 2.24^{a}	5.09 ± 3.02a	6.36 ± 4.52^{a}
DO ($mg \cdot dm^{-3}$)	10.57 ± 1.48^{a}	10.33 ± 1.84^{a}	10.40 ± 1.95^{a}	9.57 ± 1.81^{b}	9.17 ± 1.72^{b}	9.68 ± 1.91^{b}	8.64 ± 2.01^{b}	8.98 ± 1.99^{b}	8.79 ± 1.81^{b}
TH ($mg \cdot dm^{-3}$)	339.7 ± 82.5^{b}	411.73 ± 294.8^{a}	228.6 ± 33.2^{c}	316.3 ± 54.1^{b}	275.6 ± 63.7^{b}	214.8 ± 17.0^{c}	280.5 ± 25.4^{b}	170.7 ± 13.67^{d}	171.3 ± 12.84^{d}
pH	8.11 ± 0.13	8.10 ± 0.18	8.17 ± 0.18	7.98 ± 0.12	7.93 ± 0.19	8.09 ± 0.12	7.97 ± 0.15	7.99 ± 0.23	7.79 ± 0.20
EC ($\mu S \cdot cm^{-1}$)	398.1 ± 62.6	394.4 ± 73.5	394.3 ± 65.3	380.2 ± 46.5	385.7 ± 32.3	402.9 ± 49.6	397.7 ± 38.8	395.6 ± 34.03	405.1 ± 32.79
TDS ($mg \cdot dm^{-3}$)	245.6 ± 34.5^{a}	240.3 ± 35.2^{a}	243.7 ± 40.3^{a}	217.2 ± 24.8^{b}	220.7 ± 16.6^{b}	251.5 ± 25.7^{a}	233.7 ± 18.1^{b}	228.0 ± 16.8^{b}	241.0 ± 13.5^{a}
NH_4–N ($mg \cdot dm^{-3}$)	0.05 ± 0.03^{a}	0.05 ± 0.02^{a}	0.07 ± 0.04^{a}	0.17 ± 0.06^{b}	0.13 ± 0.04^{c}	0.11 ± 0.03^{c}	0.13 ± 0.06^{c}	0.13 ± 0.06c	0.10 ± 0.05^{ac}
NO_2–N ($mg \cdot dm^{-3}$)	0.02 ± 0.01^{a}	0.02 ± 0.01^{a}	0.01 ± 0.01^{b}	0.02 ± 0.01^{a}	0.01 ± 0.01^{b}	0.01 ± 0.01^{b}	0.02 ± 0.01^{ab}	0.01 ± 0.01^{a}	0.02 ± 0.01^{ab}
NO_3–N ($mg \cdot dm^{-3}$)	1.47 ± 0.59^{a}	1.46 ± 0.83^{a}	1.31 ± 0.68^{a}	1.01 ± 0.34^{b}	0.84 ± 0.24^{c}	1.05 ± 0.43^{b}	0.83 ± 0.36^{c}	0.81 ± 0.28^{c}	0.86 ± 0.26^{c}
TN ($mg \cdot dm^{-3}$)	2.17 ± 0.82^{a}	2.23 ± 0.99^{a}	1.98 ± 0.84^{a}	1.85 ± 0.16^{ab}	1.72 ± 0.06^{b}	1.63 ± 0.57^{b}	1.88 ± 0.23^{ab}	1.64 ± 1.18^{b}	1.98 ± 0.72^{a}
PO_4–P ($mg \cdot dm^{-3}$)	0.03 ± 0.01^{a}	0.03 ± 0.01^{a}	0.03 ± 0.02^{a}	0.06 ± 0.02^{b}	0.06 ± 0.02^{b}	0.04 ± 0.02^{a}	0.06 ± 0.01^{b}	0.06 ± 0.02^{b}	0.10 ± 0.03^{c}
TP ($mg \cdot dm^{-3}$)	0.12 ± 0.03^{b}	0.11 ± 0.03^{b}	0.10 ± 0.03^{a}	0.10 ± 0.04^{a}	0.08 ± 0.03^{a}	0.09 ± 0.03^{a}	0.08 ± 0.04^{a}	0.10 ± 0.07^{ab}	0.19 ± 0.05^{c}
Na^{+} ($mg \cdot dm^{-3}$)	13.44 ± 3.4^{a}	15.25 ± 4.4^{b}	13.91 ± 4.9^{a}	12.67 ± 6.2^{a}	11.60 ± 2.7^{c}	16.29 ± 2.13^{b}	12.43 ± 2.92^{a}	12.02 ± 1.94^{a}	14.70 ± 3.97^{ab}
K^{+} ($mg \cdot dm^{-3}$)	1.99 ± 0.5^{a}	1.99 ± 0.28^{a}	2.06 ± 0.36^{a}	2.10 ± 1.15^{a}	2.07 ± 1.27^{a}	2.22 ± 0.68^{b}	2.65 ± 1.10^{b}	2.28 ± 1.11^{b}	3.46 ± 1.25^{c}
Ca^{++} ($mg \cdot dm^{-3}$)	53.83 ± 7.4^{a}	53.72 ± 7.8^{a}	52.53 ± 7.6^{a}	53.01 ± 6.6^{a}	53.98 ± 6.38^{a}	54.47 ± 5.22^{a}	60.08 ± 6.94^{b}	59.94 ± 7.17^{b}	62.00 ± 7.38^{b}
Mg^{++} ($mg \cdot dm^{-3}$)	13.43 ± 3.0^{a}	11.29 ± 2.55^{b}	13.54 ± 2.73^{a}	15.98 ± 3.21^{c}	15.92 ± 5.04^{c}	12.41 ± 2.51^{ab}	13.49 ± 2.69^{a}	12.83 ± 4.18^{a}	12.85 ± 3.85^{a}
Cl^{-} ($mg \cdot dm^{-3}$)	20.64 ± 5.8	21.19 ± 7.54	21.37 ± 7.70	20.73 ± 4.26	20.41 ± 3.80	23.61 ± 6.28	20.53 ± 3.92	20.15 ± 4.65	20.50 ± 2.26
SO_4^{--} ($mg \cdot dm^{-3}$)	36.83 ± 9.4^{a}	30.18 ± 5.19^{b}	35.42 ± 9.02^{a}	27.42 ± 2.50^{bc}	25.50 ± 1.93^{c}	34.00 ± 5.51^{a}	26.75 ± 4.27^{c}	26.25 ± 3.08^{c}	27.27 ± 3.20^{c}
Fe^{+++} ($\mu g \cdot dm^{-3}$)	582.4 ± 245.9^{a}	535.3 ± 162.6^{a}	417.1 ± 330.5^{b}	488.8 ± 265.9^{ab}	225.9 ± 123.8^{c}	710.5 ± 413.2^{d}	229.8 ± 83.7^{c}	224.7 ± 113.2^{c}	406.2 ± 183.8^{b}
Mn^{++} ($\mu g \cdot dm^{-3}$)	39.9 ± 22.8^{a}	36.93 ± 11.5^{a}	29.67 ± 17.39^{a}	34.24 ± 13.99^{a}	31.40 ± 8.26^{a}	63.36 ± 50.2^{b}	22.05 ± 5.26^{c}	24.10 ± 10.7^{c}	32.37 ± 12.35^{a}
Zn^{++} ($\mu g \cdot dm^{-3}$	81.07 ± 53.2^{a}	59.5 ± 19.1^{ab}	33.18 ± 25.1^{b}	15.44 ± 8.0^{c}	9.43 ± 1.63^{c}	77.19 ± 21.72^{a}	18.00 ± 10.43^{c}	14.01 ± 4.24^{c}	19.00 ± 14.46^{c}
Cu^{++} ($\mu g \cdot dm^{-3}$)	20.83 ± 12.5^{a}	19.06 ± 19.32^{a}	5.86 ± 2.47^{b}	5.54 ± 2.53^{b}	5.27 ± 2.05^{b}	13.92 ± 8.59^{ab}	4.20 ± 1.60^{b}	4.58 ± 2.19^{b}	7.69 ± 3.61^{b}
Cr^{++} ($\mu g \cdot dm^{-3}$)	1.41 ± 0.81^{a}	1.16 ± 0.48^{b}	1.17 ± 0.58^{b}	2.00 ± 1.00^{c}	1.90 ± 1.73^{c}	2.25 ± 2.32^{c}	1.95 ± 2.11^{c}	1.25 ± 0.56^{b}	1.65 ± 0.63^{ac}
Pb^{++} ($\mu g \cdot dm^{-3}$)	1.52 ± 0.92^{a}	1.54 ± 0.71^{a}	1.00 ± 0.45^{b}	1.23 ± 0.62^{b}	0.63 ± 0.23^{c}	2.08 ± 1.92^{d}	0.67 ± 0.08^{c}	0.65 ± 0.13^{c}	1.38 ± 0.77^{ab}
Cd^{++} ($\mu g \cdot dm^{-3}$)	0.03 ± 0.03^{a}	0.03 ± 0.02^{a}	0.02 ± 0.01^{a}	0.03 ± 0.01^{a}	0.03 ± 0.01^{a}	0.05 ± 0.05^{a}	0.03 ± 0.02^{a}	0.10 ± 0.1^{b}	0.10 ± 0.19^{b}
Ni^{++} ($\mu g \cdot dm^{-3}$)	20.45 ± 34.8^{a}	15.56 ± 13.0^{a}	2.18 ± 1.64^{b}	1.63 ± 0.47^{b}	1.53 ± 0.45^{b}	8.05 ± 9.57^{ab}	2.73 ± 0.94^{b}	2.83 ± 0.77^{b}	2.52 ± 0.83^{b}
Al^{++} ($\mu g \cdot dm^{-3}$)	332.9 ± 148.3^{a}	317.2 ± 94.8^{a}	252.7 ± 185.3^{a}	299.3 ± 147.2^{a}	151.8 ± 66.8^{b}	404.5 ± 221.6^{c}	147.6 ± 44.3^{b}	174.5 ± 86.6^{b}	249.2 ± 90.5^{ab}
TOC ($mg \cdot dm^{-3}$)	4.78 ± 2.00^{a}	4.70 ± 1.01^{a}	4.03 ± 0.46^{a}	2.85 ± 0.64^{b}	3.20 ± 0.71^{b}	4.48 ± 0.76^{a}	3.45 ± 1.52^{ab}	3.20 ± 0.63^{b}	3.27 ± 0.50^{b}

Various letters in the poem indicate statistical differences between the stations.

at stations 1 and 6 (Fig. 2). Although statistically insignificant ($p > 0.05$), the WQI value was higher in the summer period than in the other seasons. The mean WQI values were 94.2, 93.4, 95.9, and 87.2 in winter, spring, summer, and autumn, respectively. These results show that the water quality was rated good in the autumn and excellent in other seasons.

3.3. Proposed WQI_{min}

Typically 9–11 physical and chemical parameters are used to evaluate WQI calculations. In developing countries with tight budgets, it is often impossible to conduct water quality surveys involving a wide range of analyses. Principal component analysis (PCA) and correlation analysis were performed for only 16 parameters. The reason for the rejection of the remaining 13 parameters was the lack of a normal distribution of the data or very low values of the factor vectors.

Significant axes were selected based on the Kaiser criterion, taking into account in the further analysis only those components (PC1, PC2, PC3, PC4, PC5) whose eigenvalues were greater than 1 (Table 4). This allowed explain almost 73% of the total variability of the data set, the initial set of parameters. Each principal component delineated a homogeneous group of primary variables. The first component of PC1 was most strongly generated by EC and TDS. In the graph, these are the longest vectors forming a small angle, which indicates a large correlation between the EC and TDS indices (Fig. 3). WT and DO were also important. The signs of the factor loadings indicate that the WT variable influences the PC1 component opposite to the EC, TDS, and DO variables. Factors Cl^-, NO_3–N, and Ca^{++} are slightly shorter vectors, proving their moderate influence on the first significant component (PC1). The first component of PC1, corresponding to the highest eigenvalue, explained 29.67% of the total variance. The second component of PC2 mainly represented the primary variable PO_4–P (strong positive correlation) and was moderately related to SS (negative correlation). Factors TH, pH, and NH_4–N are vectors slightly shorter. PC2 explained almost 19% of the total variance. The third component's dominant charge was derived from the TP parameter (negative, moderate correlation). PC3 explained more than 9% of the total variance. The fourth component of PC4 represented mainly NO_2–N (negative, moderate correlation), and the fifth variable component was Mg (negative, moderate correlation). The last two principal components accounted

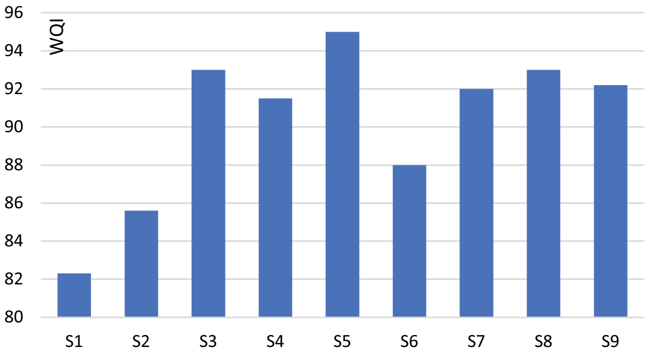

Fig. 2. Spatial variation of WQI_{full}.

for almost 15% of the total variability. Based on the PCA analysis, the following parameters are necessary to identify the water quality: EC, TDS, WT, DO, PO_4–P, and, to a lesser extent: NH_4–N, NO_3–N, Cl^-, SS, TP.

The Pearson correlation matrix (Table 5) was used to determine the relationship between 16 water quality variables. As expected, EC was highly significantly correlated with TDS and Cl^- because the higher the salt and solute concentration, the higher the conductivity [59]. High concentrations of nutrients lead to an increase in the concentration of organic matter, contributing to a decrease in DO and breathing difficulties [48,50]. The decrease in DO content is related to the increase in EC, TDS, and WT. Thus, the main sources of pollution in the river were untreated domestic sewage, sewage from fish and cattle farms, and soil erosion [56]. Based on the analysis of correlation, WT and EC were eliminated from the parameters selected based on PCA. Finally, the following parameters were selected for water quality identification: TDS, Cl^-, DO, PO_4–P, NO_3–N, SS, NO_2–N, and TP. The results of our water quality analyses in 2019 confirm the results of the 2014 research in the Danube River basin [55].

Dissolved oxygen (DO) is one of the most important parameters for assessing water quality due to its effect on living organisms in surface water. Very high or low DO levels affect water quality and can destroy aquatic life. The level of DO in natural waters depends on several factors such as aeration, temperature, photosynthesis, respiration, salinity, and atmospheric pressure. Proper functioning of life in water requires a DO concentration above 5 $mg \cdot dm^{-3}$, while a concentration below 2 $mg \cdot dm^{-3}$ can lead to the death of fish

Table 4
Matrix of factor loadings determined based on water quality parameters

Parameters	PC1	PC2	PC3	PC4	PC5
SS	0.10	**–0.72**	–0.35	–0.17	0.21
DO	**–0.81**	–0.37	0.16	0.08	0.14
TH	0.16	–0.48	0.48	–0.15	0.04
pH	–0.37	–0.56	0.05	0.38	0.29
EC	**–0.89**	0.31	–0.10	0.13	–0.06
TDS	**–0.89**	0.08	–0.27	0.06	–0.08
NH_4–N	–0.07	**0.66**	0.41	0.11	0.33
NO_2–N	–0.25	0.08	0.25	–0.51	0.37
NO_3–N	**–0.61**	–0.46	0.15	–0.37	0.04
PO_4–P	–0.03	**0.84**	–0.12	–0.35	0.01
TP	–0.07	0.18	**–0.68**	–0.36	0.35
Ca^{++}	–0.53	0.50	0.10	0.25	0.36
Mg^{++}	–0.44	0.01	0.32	–0.42	–0.54
Cl^-	**–0.69**	0.21	–0.14	0.16	–0.36
SO_4^{++}	–0.48	–0.39	–0.36	–0.07	–0.14
WT	**0.85**	0.03	–0.20	0.03	–0.15
Eigenvalue	4.75	3.02	1.48	1.27	1.13
Variance %	29.67	18.85	9.24	7.94	7.06
Cumulative variance %	29.67	48.52	57.76	65.70	72.76

Statistically significant values are shown in bold.

[60,61]. Due to its crucial importance for life, the oxygen content in water is very often used to determine the water quality index. The situation is different in the case of water temperature, which, being a climatic variable, is often ignored. Many studies show a negative correlation between DO and WT [50,62,63]. The water temperature was significantly lower in the winter season than in the summer season ($p < 0.01$). Temperature drop and inhibition of vegetation in the winter season very often led to increased water oxygenation. The increase in temperature in the summer period sometimes contributes to a reduction of the river's water flow and algal blooms' occurrence [64]. Lowering the water level and limiting light access causes a decrease in oxygen content [65]. There was also a high negative correlation between WT, EC, and TDS. The increase in the concentration of EC and TDS in the winter season may result from the increased demand for energy produced from non-renewable sources [66].

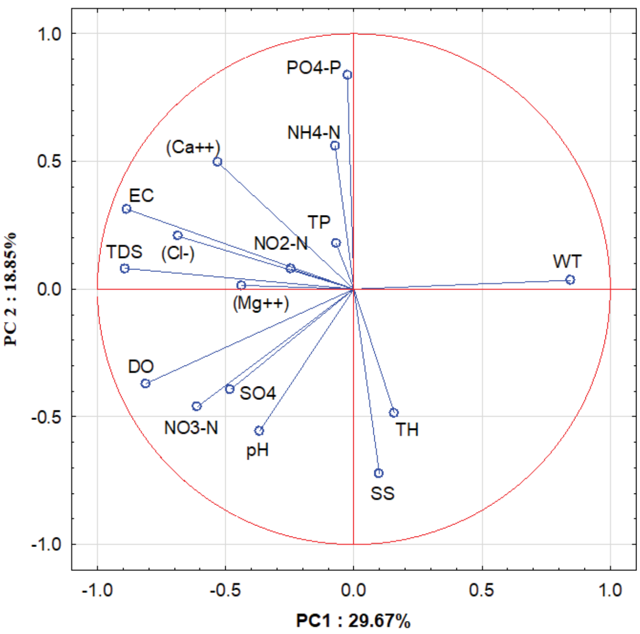

Fig. 3. PCA circle graph.

The presence of nutrients in rivers that are drinking water sources potentially threatens people's health [67]. The nutrient content in this study was significantly higher in the winter than in the summer. High SS content is associated with river siltation and plant rot, which causes an increase in EC, contributing to the deterioration of water quality [68]. A high inverse correlation was found between SS and PO_4–P concentrations. The highest concentrations of SS were observed in the spring at S6. High values of SS concentrations were related to the occurrence of rainfall runoff from the airtight surfaces of industrial plants. On the other hand, the concentrations of PO_4–P showed no statistically significant seasonal variation. Research conducted in the catchments of the Tama, Bystrzyca, and Ochta rivers showed a significant influence of urbanized areas on the increase of SS and TDS concentrations [49,69,70].

3.4. Water quality assessment using WQI_{min}

The results from WQI_{min} showed about 9 (annual average) lower water quality in stations located in the upper and middle sections of the river (S1, S2, S6). This was in line with the changing water quality trend predicted by WQI_{full}. The lower water quality at stations S1 and S2 resulted from the inflow of pollutants from the neighboring countries (Fig. 4). The cause of the river pollution in this area is the inflow of untreated municipal and industrial wastewater from the city of Pécs (Hungary) [71]. On the other hand, the decrease

Table 5
Correlation matrix for important water quality parameters

Parameters	SS	DO	TH	pH	EC	TDS	NH_4–N	NO_2–N	NO_3–N	PO_4–P	TP	Ca^{++}	Mg^{++}	Cl^-	SO_4^{++}	WT
SS	1.00															
DO	0.16	1.00														
TH	0.20	0.06	1.00													
pH	0.29	–0.56	0.17	1.00												
EC	–0.31	–0.57	–0.29	0.20	1.00											
TDS	0.58	–0.58	–0.26	0.23	**0.88**	1.00										
NH_4–N	–0.38	–0.07	–0.10	–0.10	0.17	0.02	1.00									
NO_2–N	–0.02	0.12	0.01	–0.03	0.17	0.15	0.15	1.00								
NO_3–N	0.26	0.61	0.20	0.31	0.30	0.44	–0.20	0.33	1.00							
PO_4–P	–0.49	–0.29	–0.32	–0.52	0.25	0.06	0.35	0.15	–0.24	1.00						
TP	0.18	–0.04	–0.19	–0.04	0.12	0.17	–0.04	0.03	–0.04	0.39	1.00					
Ca^{++}	–0.41	0.27	–0.25	0.02	0.57	0.48	0.37	0.16	0.11	0.33	0.02	1.00				
Mg^{++}	–0.15	0.34	0.05	–0.01	0.30	0.28	0.05	0.14	0.35	0.14	–0.09	–0.05	1.00			
Cl^-	–0.16	0.36	–0.20	0.13	**0.77**	**0.70**	0.05	0.02	0.23	0.14	0.01	0.31	0.32	1.00		
SO_4^{++}	0.23	0.36	–0.03	0.24	0.29	0.56	–0.26	0.06	0.44	–0.29	0.06	0.04	0.14	0.20	1.00	
WT	0.08	**–0.87**	0.04	–0.28	–0.48	–0.63	–0.12	–0.19	–0.53	0.00	–0.05	–0.47	–0.40	–0.38	–0.25	1.00

Statistically significant values are shown in bold.

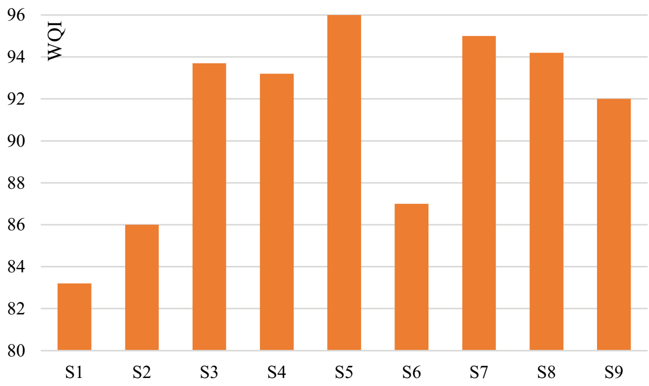

Fig. 4. Spatial variation of WQI_{min}.

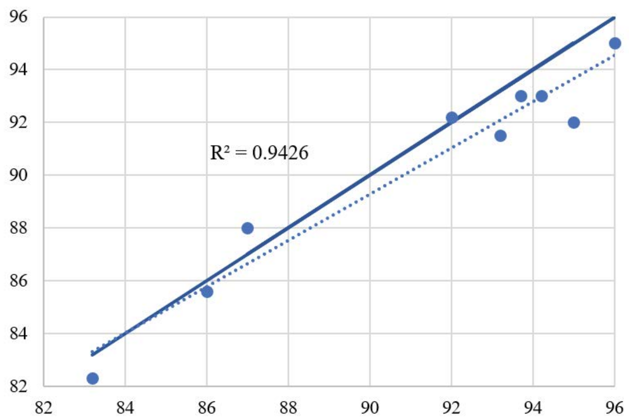

Fig. 5. Correlation between WQI_{full} and WQI_{min}.

in water quality at the S6 station resulted from the discharge of industrial and fish farm sewage.

The spatial analysis showed that the WQI_{full} values were very close to the WQI_{min} (Fig. 5). Temporal analysis showed that WQI_{full} values were the lowest in autumn, while when applied, they were lowest in winter. Significantly lower WQI_{min} values in the winter season result from higher concentrations of NO_3–N and SS and higher DO, as well as higher weights of these parameters. Despite the differences between the indicators, the WQI_{min} can be used for a simplified analysis of changes in time and space in water quality. A simplified procedure is intended to reduce the scope and costs of chemical analyses [72].

The spatial trends in water quality changes are the same as in the case of the Bug River in Poland. In both cases, the Danube and Bug rivers, the highest pollution was found in the upper section of the river. The highest water quality was observed in summer and the lowest in winter [50]. The leading cause of increased pollution in winter is the increase in energy demand and the limitation of vegetation.

4. Conclusion

A tool in the form of WQI was used to assess the spatial variability and classification of drinking water quality. The highest pollution level was found at stations located in the upper section of the river (Bezdan, Bogojevo) and one station in the middle section (Banatska Palanka). The lower quality of water in the upper section of the river results from the inflow of pollutants from catchments located in other countries. Discharging industrial wastewater and sewage from fish farms and soil erosion are responsible for increasing pollution in the middle section. Based on the PCA and correlation analysis, some water quality parameters were eliminated, and a modified WQI_{min} was used for the evaluation. Reducing the number of analyzed parameters contributes to the reduction of the costs of the analysis without losing the correctness of the obtained results. Modified WQI_{min} can be used for future monitoring of the Danube River water quality in other countries. Proper management of water resources is possible by implementing water pollution prevention strategies as part of international cooperation. The essential element of collaboration is the European Strategy for the Danube Region.

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