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A Comparative Approach to a Series of Physico-Chemical Quality Indices Used in Assessing Water Quality in the Lower Danube

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Abstract: Water quality indices are suitable tools used for assessing water quality because of their capacity to reduce a large number of water quality indicators into one value which defines the water quality class. In this study, Water Quality Index (WQI), Water Pollution Index (WPI) and Canadian Council of Ministers of the Environment Water Quality Index (CCME-WQI) were applied in order to evaluate the seasonal and spatial variation of the water quality in the Romanian Lower Danube sector. Fourteen physico-chemical parameters, i.e., pH, DO, BOD_5, COD, $N\text{-}NH_4^+$, $N\text{-}NO_3^-$, $N\text{-}NO_2^-$, N-total, P-total, SO_4^{2-}, Cl^-, Fe-total, Zn^{2+} and Cr-total, were monitored along the Danube course (on a distance of about 120 km), during the four seasons between the autumn of 2018 and the summer of 2019 in order to calculate the three indices mentioned above. Indices results showed that the water analysed was ranked into different water quality classes, although the same dataset was used. These differences were due to the contribution of each parameter taken into account in the calculation formula. Thus, the WQI scores were mostly influenced by those parameters whose maximum allowable concentration was low (e.g., heavy metals, $N\text{-}NO_2^-$), while the WPI and CCME-WQI scores were influenced by those parameters which exceeded the maximum allowable concentration (BOD_5, DO, COD, $N\text{-}NO_3^-$, $N\text{-}NO_2^-$). Based on the WQI results, the water was ranked into quality classes II and III. WPI and CCME-WQI assessed water only in quality class II, with one exception in the case of CCME-WQI when water was ranked into quality class III. The temporal assessment identified the seasons in which the water quality was lower, namely summer and autumn. The variation of the indices values between the sampling stations demonstrates the existence of pollution sources in the study area. Moreover, the indices results illustrated the contribution of the main tributaries (Rivers Siret and Prut) to the Danube River water quality. The appropriate applicability of the three indices was also discussed in this study.

Keywords: water quality indices; Lower Danube; physico-chemical parameters; quality class; comparative analysis

1. Introduction

Permanent water quality monitoring and assessment of surface water quality are indispensable for preserving the global ecosystem and human health [1]. Thus, once the state of water quality is known, the next step is to find solutions to reduce pollution and rational management of water resources [2–7].

Water quality indices are helpful tools used for water quality assessment. They have been used since 1965, when Horton developed the first form of the Water Quality Index (WQI) [8]. The main aim of quality indices is to reduce a large number of physical, chemical and biological measurements to a single value which indicates the ecological status of a specific watercourse [9–12]. The general algorithm for calculating water quality indices consists in converting all the parameters values to a common scale (sub-indices) and then, afterwards, aggregating them into one final value (index). The index value is used to establish the water quality status and, sometimes, specific water uses (drinking, bathing, irrigation, recreation, domestic and industrial purposes) [13,14].

Most water quality indices, such as the Water Quality Index (WQI), the Canadian Council of Ministers of the Environment Water Quality Index (CCME-WQI), the Prati's Implicit Index of Pollution, the Universal Water Quality Index, the British Columbia Water Quality Index (BCWQI), the Oregon Water Quality Index (OWQI), the Water Pollution Index (WPI), the Serbian Water Quality Index (SWQI), the Overall Index of Pollution (OIP), etc., are based on physico-chemical parameters and seldom faecal coliforms bacteria [15,16]. Apart from physico-chemical indices, various biotic indices, such as the Saprobic Index, the Shannon Index, Pielou's Index, the Biological Monitoring Working Party (BMWP), the Trent Biotic Index (TBI), the Belgian Biotic Index (BBI), Macroinvertebrate Community Index (MCI) and the Danish Stream Fauna Index (DSFI) [17], are used for the bioassessment of aquatic ecosystems. A complex and accurate evaluation of an aquatic ecosystem quality implies applying both physico-chemical and biological indices [18].

In order to assess water quality in the Danube River from a physico-chemical point of view, various indices have been calculated, such as Water Quality Index (WQI) [19], Canadian Council of Ministers of the Environment Water Quality Index (CCME-WQI), [20,21], Water Pollution Index (WPI) [22], Serbian Water Quality Index [23–25], Bascaron Water Quality Index (BWQI) [26] and Heavy Metal Pollution Index [27]. Based on the results obtained, the Danube water was classified into different quality classes. For example, the Danube water in Romania was mostly evaluated by the WQI as "good" quality water (class II) [19], with the same water being "marginal" and "fair" according to the CCME-WQI and "good" according to the BWQI [26]. In the assessment of the Serbian area of the Danube, CCME-WQI, SWQI and WPI were applied. SWQI results overall assessed the water quality as "good" and "very good", while CCME-WQI results assessed it as "fair" and "marginal" [23]. According to the WPI scores, water quality was mainly "moderately polluted" and "polluted" [22].

Taking into account the aspects mentioned above, the present paper aims at testing and comparing the applicability of Water Quality Index, Water Pollution Index and of the Canadian Council of Ministers of the Environment Water Quality Index in order to obtain an accurate representation of water quality in the studied area, namely the Romanian Lower Danube sector bordering the cities of Galati, Braila and Tulcea. In order to reach this aim, a common dataset was used to calculate the three indices and, based on the results obtained, the water quality was included in the quality classes specific to each index. Moreover, the parameters which had the highest weight in the calculation formula of the indices were established.

The Danube sector assessed in the present study covers the confluence area of the Lower Danube with two major rivers, Siret and Prut, and the predeltaic Danube Area, which represents the transport route of pollutants to the one of the most important natural reservations in the world, namely the Danube Delta Biosphere Reserve. Half of the analysed area (the sector between sampling stations P8–P15) represents a Site of Community Importance (ROSCI0065) [28] and a Special Bird Protection Area (ROSPA0031) [29]. These two Natura 2000 sites shelter a wide variety of rare, threatened or endemic flora and fauna species, and various habitats protected by the Habitats Directive and Birds Directive [30,31]. Thus, the area must be constantly monitored in order to create a habitat conducive to protected species.

2. Materials and Methods

2.1. Sampling and Analysis Procedures

In order to carry out this study, samples were taken from 15 sampling stations during the four seasons between the autumn of 2018 and the summer of 2019. Water samples were collected monthly, except for the winter season, which allowed for water sampling only in December, due to weather conditions (water freezing). More precisely, 10 water samples were taken from each station, with the total number of samples being 150. Fourteen physico-chemical parameters were measured both in-situ and ex-situ using standardised methods, as presented in Table 1. The samples analysed ex-situ were taken in polyethylene (PE) containers and were analysed in CREDENTIAL, a laboratory which is part of the European Center of Excellence for the Environment (ECEE) of "Dunarea de Jos" University of Galati.

Table 1. Physico-chemical parameters and reference methods used [32].

	Parameter (Abbreviation)	**Reference Method**
in-situ	pH	SR EN ISO 10523:2012
	Dissolved Oxygen (DO)	SR ISO 5814:1984
	Biochemical oxygen demand (BOD_5)	SR EN 1899-2:2002
ex-situ	Chemical oxygen demand (COD)	SR ISO 15705:2002
	Ammonium nitrogen ($N\text{-}NH_4^+$)	SR EN ISO 11732:2005
	Nitrate nitrogen ($N\text{-}NO_3^-$)	SR EN ISO 11905-1:2003
	Nitrite nitrogen ($N\text{-}NO_2^-$)	SR EN 26777:2006
	Total nitrogen (N-total)	SR EN ISO 11905-1:2003
	Total phosphorus (P-total)	SR EN ISO 6878:2005
	Ion sulphate (SO_4^{2-})	STAS 3069-87
	Ion chloride (Cl^-)	SR ISO 9297/2001
	Total iron (Fe-total)	SR ISO 6332:1996/C91:2006
	Total chrome (Cr-total)	SR ISO 9174-98
	Zinc (Zn^{2+})	Photometric method—reaction of alkaline solution zinc ions with pyridylazoresorcinol

2.2. *Study Area*

The water samples were collected from 15 stations located on a 120 km sector along the Lower Danube, the first station being upstream Braila (P1) and the last one downstream Tulcea (P15) (Figure 1). The stations were established in such a way as to include areas sensitive to pollution generated by anthropogenic factors (P2, P3, P6, P9, P10, P13, P14, P15), but also areas where the anthropogenic influence is minimal (P4, P5, P8). Agricultural, industrial and domestic activities are the main sources of pollution in the study area [33]. Another important source is represented by the treated waters discharged from the existing Wastewater Treatment Plants (WWTP) in the three cities, namely Braila, Galati and Tulcea, bordering the monitored sector. More precisely, the stations prone to permanent pollution are located as follows:

- P2 and P14 near two shipyards.
- P3 in the ferry crossing area (Braila City).
- P10 and P13 in the vicinity of agricultural lands.
- P6 at the confluence of the Danube with River Siret. This tributary is the emissary of treated waters coming from the municipal wastewater treatment plant (Galati City) and from the sewage treatment plant of an important steel mill.
- P9 at the confluence of the Danube with River Prut. This river crosses a large area where agricultural and industrial activities are carried out intensively and it represents the natural border between

Romania and the Republic of Moldova. This aspect is relevant considering the fact that wastewater is discharged in River Prut from the territory of this country, as well.

- P15 in the vicinity of the urban agglomeration of Tulcea City.

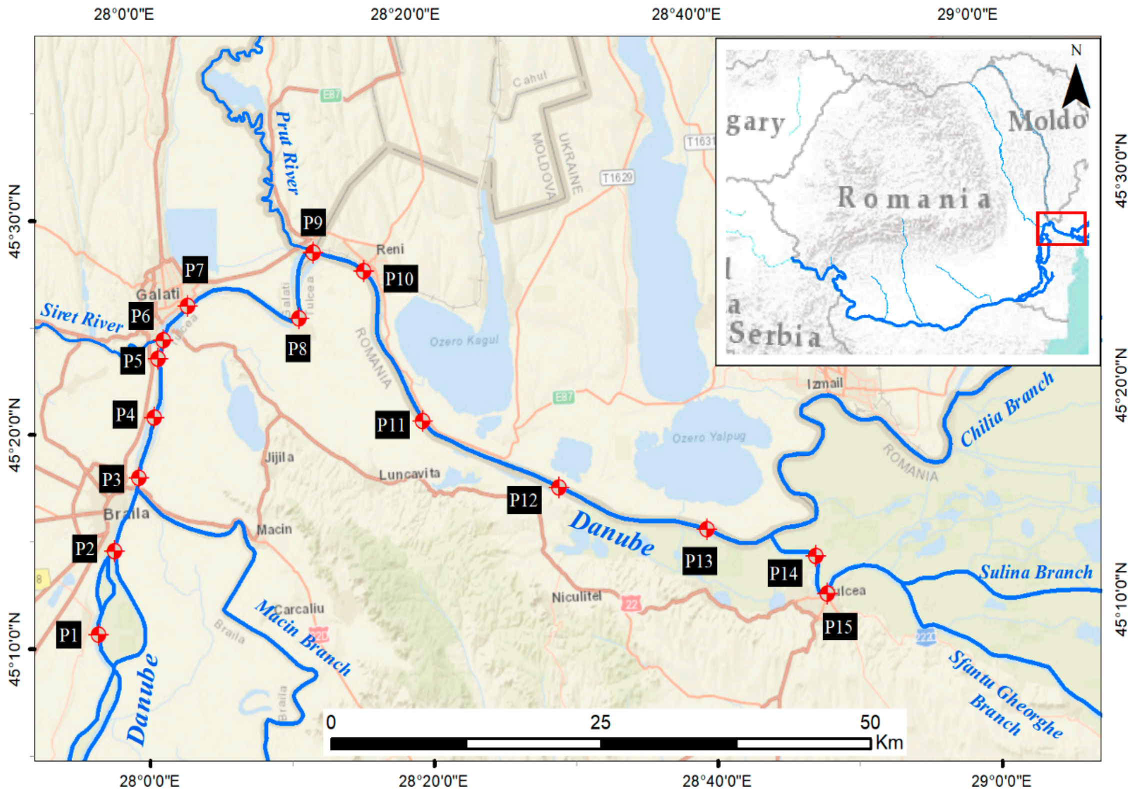

Figure 1. Distribution of the sampling points in the Romanian Lower Danube study area.

The pluviometric regime and the climatic conditions influence the Danube water flows depending on the seasons in which the studies are carried out. Thus, the average annual temperature is around 10 °C, and the average summer temperature is 21.3 °C. The rainy periods are usually in spring, and the periods of prolonged drought extend over a fairly long period of time, between August and November. For this reason, the highest flows are between April and June, and the lowest are recorded in September–October. Moreover, the main tributaries of the Danube in the studied area (Rivers Siret and Prut) are under the same climatic and pluviometric regime, respectively.

2.3. Data Analyses

The differences in the calculation algorithms of the three indices, especially in the way of aggregating the parameters, was the main reason for choosing to apply and compare them in the present study. Moreover, these indices allowed the use of a common set of water quality parameters, an essential aspect for obtaining an accurate comparison. Thus, the same dataset was used in the calculation formulas of each index.

- Water Quality Index (WQI)

The weighted Arithmetic Water Quality Index method was used in the present paper. The WQI was calculated by using the following equation [34–37]:

$$\mathrm{WQI} = \frac{\sum \mathrm{Wi} \times \mathrm{qi}}{\sum \mathrm{Wi}} \quad (1)$$

where, Wi represents the relative weight of each parameter i taken into consideration and qi represents the quality rating scale for each parameter i, qi is calculated based on the following variables: the experimentally determined value of parameter i, the ideal value of this parameter (it is 0 for all the parameters except for the pH whose value is 7 and the DO whose value is 14.6 $mg \cdot L^{-1}$) and the maximum allowed concentration (MAC) regulated by Order 161/2006, submitted for quality class II (Table 2).

Table 2. Maximum allowed concentration of each parameter regulated by Order 161/2006 [38].

Parameter	Unit	Maximum Allowed Concentration ($mg \cdot L^{-1}$)
pH	upH	8.2
DO	$mg\ O_2 \cdot L^{-1}$	>7
BOD_5	$mg\ O_2 \cdot L^{-1}$	5
COD	$mg\ O_2 \cdot L^{-1}$	25
$N\text{-}NH_4^+$	$mg\ N \cdot L^{-1}$	0.8
$N\text{-}NO_3^-$	$mg\ N \cdot L^{-1}$	3
$N\text{-}NO_2^-$	$mg\ N \cdot L^{-1}$	0.03
SO_4^{2-}	$mg \cdot L^{-1}$	120
Cl^-	$mg \cdot L^{-1}$	50
N-total	$mg \cdot L^{-1}$	7
P-total	$mg \cdot L^{-1}$	0.4
Fe-total	$mg \cdot L^{-1}$	0.5
Zn^{2+}	$mg \cdot L^{-1}$	0.2
Cr-total	$mg \cdot L^{-1}$	0.05

Based on the WQI value, surface water can be classified into five quality classes, as described in Table 3.

Table 3. Water quality status based on Water Quality Index (WQI).

WQI Values	Status
0–25	Excellent (I)
26–50	Good (II)
51–75	Poor (III)
76–100	Very poor (IV)
>100	Unsuitable for drinking (V)

- Water Pollution Index (WPI)

Calculation of the Water Pollution Index was carried out by using Equation (2). The experimental value of each parameter (Ai) was divided by the maximum concentration allowed (T) by national legislation (Order 161/2006). Afterwards, the sum of the results obtained was calculated and divided by the number of parameters (n = 14) used. The water quality classification according to WPI is shown in Table 4 [22,39,40].

$$\mathrm{WPI} = \frac{1}{\mathrm{n}} \sum_{\mathrm{n}=1}^{\mathrm{n}} \frac{\mathrm{Ai}}{\mathrm{T}} \tag{2}$$

Table 4. Water quality classification based on the Water Pollution Index (WPI).

WPI Value	Water Quality Class
≤0.3	I—Very pure
0.3–1.0	II—Pure
1.0–2.0	III—Moderately polluted
2.0–4.0	IV—Polluted
4.0–6.0	V—Impure
≥6.0	VI—Heavily impure

- Canadian Council of Ministers of the Environment Water Quality Index (CCME-WQI)

The Canadian Council of Ministers of the Environment Water Quality Index (CCME-WQI), also known as the Canadian Water Quality Index (CWQI), presents a calculation algorithm different from the other two indices applied in this study. The computation method (Equation (3)) is based on three main factors (F_1, F_2, F_3), which describe: Scope—the percentage of indicators which do not meet water quality objectives (F_1), Frequency—the number of times the indicators exceeded the threshold value (F_2) and Amplitude—the extent to which the indicators' concentrations are greater (or lower for dissolved oxygen) than the threshold value (F_3) [41–45]. The calculation formulas of the three factors are detailed in the Canadian Water Quality Guidelines for the Protection of Aquatic Life [46].

$$\text{CCME-WQI} = 100 - \left(\frac{\sqrt{F1^2 + F2^2 + F3^2}}{1.732}\right) \quad (3)$$

Based on the CCME-WQI, surface waters are classified into 5 quality categories, which are summarized in Table 5.

Table 5. Water quality category according to the Canadian Council of Ministers of the Environment Water Quality Index (CCME-WQI).

CWQI Value	Quality Category
95–100	Excellent (I)
80–94	Good (II)
65–79	Fair (III)
45–64	Marginal (IV)
0–44	Poor (V)

3. Results and Discussion

3.1. Water Quality Assessment Using WQI, WPI and CCME-WQI Indices

The preliminary statistical analysis of the experimentally determined values for each parameter monitored in the 15 sampling stations is presented in Table 6. Based on this dataset, the three indices were calculated in order to assess and classify the water quality.

Table 6. Preliminary statistical analysis of the experimental dataset.

		P1	P2	P3	P4	P5	P6	P7	P8	P9	P10	P11	P12	P13	P14	P15
pH (upH)	Mean	7.14	7.43	7.64	7.53	7.96	7.92	8.12	7.61	8.24	7.73	7.86	7.86	7.84	7.68	7.49
	Min	6.86	7.23	7.45	7.04	7.40	7.55	7.31	6.50	7.00	7.28	7.55	7.50	7.47	7.33	7.17
	Max	7.97	7.88	7.88	7.87	8.50	8.32	9.37	8.22	10.02	8.27	8.23	8.21	8.16	8.13	8.19
	Std Dev	0.38	0.23	0.16	0.27	0.37	0,29	0.83	0.72	1.18	0.43	0.29	0.30	0.29	0.37	0.34
DO ($mg{\cdot}L^{-1}$)	Mean	9.60	9.50	9.75	6.01	8.49	8.34	8.99	8.83	8.86	9.27	9.10	9.41	10.21	9.55	8.75
	Min	6.58	6.69	6.40	3.20	3.26	3.26	3.13	3.68	3.72	7.25	5.96	6.09	6.28	6.36	4.83
	Max	12.63	12.92	12.76	12.10	12.86	13.16	13.00	13.77	12.48	11.36	11.78	12.94	13.29	11.61	11.09
	Std Dev	2.73	2.71	2.83	3.41	3.84	3.78	3.50	3.96	3.23	1.96	2.55	2.94	3.32	2.53	2.93
BOD_5 ($mg{\cdot}L^{-1}$)	Mean	7.90	8.34	7.63	6.01	8.71	9.64	7.75	8.44	11.79	4.63	7.59	6.83	8.93	6.19	6.69
	Min	1.00	1.70	2.10	3.20	0.50	1.00	1.60	4.00	4.00	1.00	1.60	3.40	4.00	3.50	1.00
	Max	17.00	15.90	14.80	18.10	15.90	20.80	15.00	13.10	19.20	15.00	12.00	10.30	15.30	7.60	10.90
	Std Dev	5.82	6.09	5.65	5.41	6.11	8.00	4.64	3.71	6.57	5.59	4.39	3.42	3.34	1.88	4.45
COD ($mg{\cdot}L^{-1}$)	Mean	11.10	14.63	6.90	8.27	11.44	12.03	13.21	14.34	15.16	8.00	10.91	12.27	7.37	6.20	6.99
	Min	4.00	3.00	4.00	3.00	4.00	4.00	3.00	5.00	6.00	4.00	4.00	4.00	4.00	2.00	3.00
	Max	32.00	35.00	16.30	16.90	19.33	23.40	28.00	22.00	27.00	12.00	30.00	38.00	16.00	12.00	16.00
	Std Dev	10.18	11.50	4.70	5.56	6.52	9.02	9.51	6.59	8.05	3.11	8.92	10.96	4.00	3.58	4.48
$N\text{-}NH_4^+$ ($mg{\cdot}L^{-1}$)	Mean	0.43	0.35	0.28	0.27	0.34	0.64	0.27	0.25	0.23	0.31	0.38	0.23	0.25	0.23	0.53
	Min	0.26	0.09	0.16	0.10	0.11	0.22	0.18	0.12	0.09	0.05	0.10	0.07	0.05	0.08	0.18
	Max	0.69	0.90	0.43	0.42	0.78	2.34	0.46	0.45	0.49	0.63	1.17	0.41	0.39	0.34	2.12
	Std Dev	0.14	0.27	0.09	0.12	0.21	0.72	0.10	0.12	0.14	0.24	0.37	0.10	0.12	0.08	0.71
$N\text{-}NO_3^-$ ($mg{\cdot}L^{-1}$)	Mean	1.77	1.67	2.41	2.34	1.59	2.13	1.63	1.65	2.40	3.24	2.67	1.86	2.09	1.89	1.97
	Min	0.80	0.80	0.80	0.70	0.70	0.90	0.70	0.80	0.90	1.70	1.50	1.40	1.40	1.60	1.40
	Max	2.60	2.60	4.70	4.60	2.80	4.40	3.50	2.70	3.40	5.60	4.90	3.30	3.10	2.20	2.70
	Std Dev	0.67	0.73	1.32	1.36	0.81	1.17	1.10	0.80	0.91	1.69	1.53	0.66	0.62	0.23	0.39
$N\text{-}NO_2^-$ ($mg{\cdot}L^{-1}$)	Mean	0.018	0.018	0.018	0.018	0.019	0.019	0.018	0.018	0.019	0.018	0.019	0.018	0.019	0.018	0.018
	Min	0.012	0.011	0.011	0.011	0.012	0.013	0.015	0.014	0.015	0.015	0.016	0.015	0.015	0.015	0.014
	Max	0.025	0.026	0.036	0.030	0.036	0.027	0.029	0.029	0.029	0.028	0.033	0.025	0.028	0.032	0.032
	Std Dev	0.004	0.004	0.008	0.006	0.007	0.005	0.005	0.005	0.004	0.004	0.006	0.003	0.004	0.006	0.006
SO_4^{2-} ($mg{\cdot}L^{-1}$)	Mean	34.57	33.71	33.43	34.57	32.50	43.75	35.25	32.88	34.00	36.00	36.86	36.14	36.14	38.43	37.43
	Min	29.00	29.00	28.00	28.00	29.00	25.00	26.00	28.00	27.00	29.00	29.00	30.00	31.00	31.00	31.00
	Max	40.00	41.00	38.00	39.00	38.00	65.00	50.00	41.00	50.00	48.00	47.00	44.00	42.00	46.00	49.00
	Std Dev	4.35	5.53	3.74	4.12	3.51	15.28	7.38	5.22	7.37	6.88	6.52	5.21	4.49	6.29	6.90

Table 6. *Cont.*

		P1	P2	P3	P4	P5	P6	P7	P8	P9	P10	P11	P12	P13	P14	P15
Cl^- ($mg \cdot L^{-1}$)	Mean	35.11	35.26	34.40	35.54	34.18	35.24	35.24	37.36	36.05	35.26	34.12	35.40	34.12	34.97	34.83
	Min	34.90	34.90	31.41	34.90	30.00	30.00	33.00	34.90	35.00	34.90	31.41	34.90	31.41	34.90	34.00
	Max	36.00	37.00	38.00	39.00	40.00	45.00	39.00	45.00	39.00	37.00	36.00	38.00	36.00	35.00	35.00
	Std Dev	0.39	0.77	2.32	1.53	3.08	4.37	1.67	4.15	1.68	0.77	1.89	1.15	1.89	0.05	0.37
N-total ($mg \cdot L^{-1}$)	Mean	1.87	1.81	1.71	1.79	1.74	2.05	2.13	1.69	1.70	1.81	1.17	1.39	1.66	1.49	1.27
	Min	0.10	0.50	0.50	0.50	0.50	0.50	0.30	0.30	0.40	0.30	0.40	0.40	0.30	0.30	0.20
	Max	3.10	3.50	3.00	4.00	2.70	3.90	4.00	2.80	4.40	6.10	2.70	2.80	5.10	3.30	2.80
	Std Dev	1.42	1.38	1.15	1.36	0.95	1.38	1.33	1.03	1.40	2.11	1.06	1.09	1.73	1.32	1.13
P-total ($mg \cdot L^{-1}$)	Mean	0.12	0.13	0.13	0.14	0.14	0.15	0.11	0.11	0.14	0.16	0.17	0.12	0.13	0.15	0.22
	Min	0.10	0.10	0.10	0.10	0.08	0.10	0.09	0.02	0.10	0.10	0.10	0.08	0.10	0.10	0.08
	Max	0.18	0.22	0.20	0.20	0.20	0.20	0.18	0.20	0.26	0.40	0.40	0.22	0.20	0.30	0.80
	Std Dev	0.03	0.05	0.05	0.04	0.04	0.04	0.03	0.06	0.06	0.11	0.11	0.06	0.05	0.07	0.26
Fe-total ($mg \cdot L^{-1}$)	Mean	0.06	0.10	0.13	0.08	0.05	0.11	0.12	0.05	0.07	0.07	0.06	0.05	0.06	0.06	0.15
	Min	0.01	0.01	0.01	0.02	0.01	0.04	0.01	0.01	0.01	0.02	0.01	0.01	0.02	0.01	0.02
	Max	0.14	0.17	0.40	0.16	0.10	0.21	0.36	0.10	0.14	0.14	0.12	0.10	0.12	0.14	0.57
	Std Dev	0.06	0.07	0.14	0.06	0.04	0.07	0.15	0.04	0.05	0.05	0.05	0.04	0.04	0.06	0.20
Zn^{2+} ($mg \cdot L^{-1}$)	Mean	0.109	0.099	0.094	0.100	0.103	0.073	0.084	0.117	0.064	0.099	0.085	0.113	0.105	0.056	0.059
	Min	0.066	0.047	0.063	0.050	0.050	0.060	0.015	0.004	0.040	0.001	0.012	0.013	0.014	0.020	0.017
	Max	0.179	0.184	0.120	0.157	0.170	0.120	0.151	0.210	0.117	0.181	0.146	0.247	0.292	0.084	0.139
	Std Dev	0.039	0.046	0.017	0.041	0.046	0.022	0.051	0.077	0.029	0.065	0.052	0.085	0.090	0.023	0.038
Cr-total ($mg \cdot L^{-1}$)	Mean	0.027	0.020	0.033	0.024	0.024	0.028	0.015	0.014	0.025	0.023	0.027	0.023	0.029	0.026	0.024
	Min	0.010	0.010	0.010	0.010	0.010	0.010	0.010	0.010	0.010	0.010	0.010	0.010	0.010	0.010	0.010
	Max	0.060	0.040	0.050	0.040	0.040	0.050	0.040	0.020	0.040	0.030	0.060	0.050	0.060	0.060	0.060
	Std Dev	0.017	0.010	0.017	0.011	0.011	0.015	0.011	0.005	0.011	0.008	0.024	0.017	0.020	0.021	0.018

The spatial and seasonal variations of the WQI, WPI and CCME-WQI results are represented in the form of distribution maps in Figures 2 and 3. A significant spatial variation was observed during summer and autumn, while there were no remarkable differences in winter and spring. Analysing the results obtained for each season, high values of the WQI and the WPI, and respectively, low values of CCME-WQI, were recorded during summer and early autumn. These results were generally influenced by BOD_5 and DO parameters, which exceeded the maximum value accepted in most sampling stations. Furthermore, both parameters were among the variables with the highest standard deviation (Table 6), which indicates an increased variability of the values, in this case a seasonal variation. The low level of dissolved oxygen which respectively increased in the case of the biochemical oxygen demand, recorded in summer and early autumn, may be explained by their dependence relation with the water temperature variation. It was found that the dissolved oxygen level decreases in the hot season, while the biological activity increases [47,48]. The seasonal variation may also be due to flow variation in the watercourse, with a high flow rate favouring the dilution of the pollutants concentration [32].

The values of the WQI varied between 40.7 and 72.5 (Figure 2), with these results ranking the water quality in classes II and III. The WQI reached its maximum value at station P14 during autumn. This result may be due to the industrial activity (shipyard) carried out in the vicinity of this monitoring station. Values over 50, which include water quality in the category "poor" (equivalent to class III), were recorded at stations P6 and P9 in all 4 seasons. These sampling stations are situated at the confluence of the Danube with its main tributaries, namely Siret (P6) and Prut (P9). Both rivers have a significant intake of pollutants, especially nutrients which originate in the fertilizers used in agricultural activities. Furthermore, River Siret is the emissary where treated domestic and industrial wastewater of Galati city is discharged.

The Water Pollution Index results illustrated in Figure 3 also highlight the contribution of the two tributaries on the quality of the Danube water. This is demonstrated by the highest WPI value recorded at station P9 (0.78) in autumn. Moreover, during spring, the highest value was obtained at station P6, when $N\text{-}NH_4^+$ and $N\text{-}NO_3^-$ exceeded maximum accepted values. However, according to the WPI scores, the water in these stations was ranked in quality class II, whereas the WQI ranked water quality in class III. High values were also recorded at P10, P11, P12 and P13 during summer and autumn, when BOD, DO, COD, $N\text{-}NO_3^-$ and $N\text{-}NO_2^-$ exceeded the acceptable limit values. These sampling stations are located in the proximity of areas with intense agricultural activity, which represents the main source of pollution. Although the values of WPI varied both spatially and temporally (the minimum value was 0.48 and the maximum, 0.78), the water was classified exclusively in quality class II ("Pure water").

Similar to the WPI, the CCME-WQI classified the water quality on the monitoring sector in class II ("Good status"), with one exception, namely the value recorded in station P11 in summer (76.54) which corresponds to quality class III ("Fair status"). This particular case is due to the fact that in the station and season mentioned above, 5 of the 14 analysed parameters (BOD, $N\text{-}NO_2^-$, $N\text{-}NO_3^-$, DO and COD) exceeded the maximum allowed concentration. Although the exceedances recorded were not significant, this result was influenced by the factor F_1—Scope indicating the percentage of variables which did not meet their objectives. Like the WQI and the WPI, the spatial distribution of CCME-WQI scores (Figure 4) and the values recorded at stations P6 and P9 strengthen the statement that the two tributaries influence the quality of the Danube water. Moreover, the CCME-WQI also captured the seasonal trends in water quality, with the values registered in summer and autumn ranking the Danube water in the lower quality class.

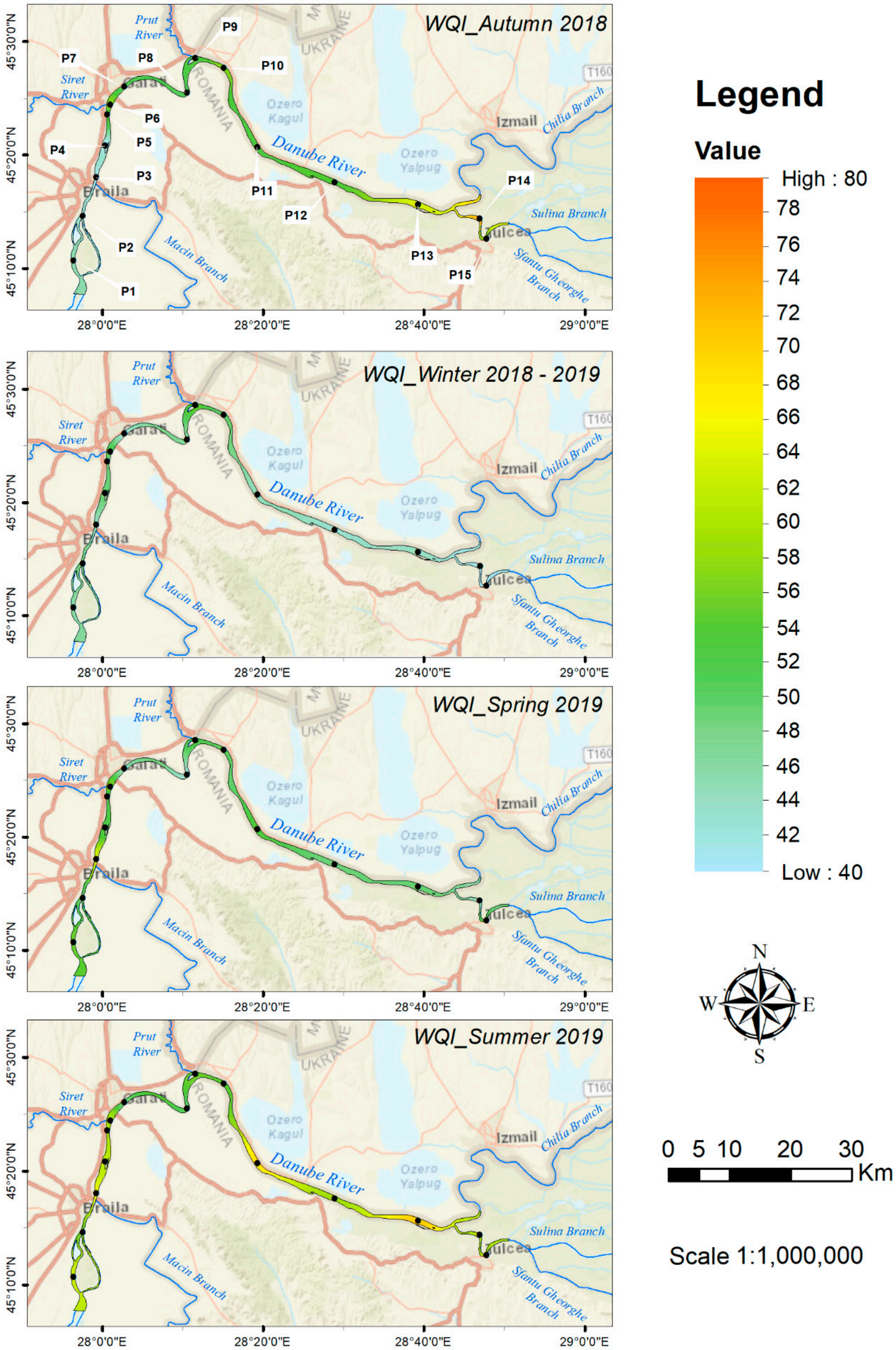

Figure 2. Spatial and temporal distribution of the WQI scores in the Romanian Lower Danube sector recorded during the period autumn 2018–summer 2019.

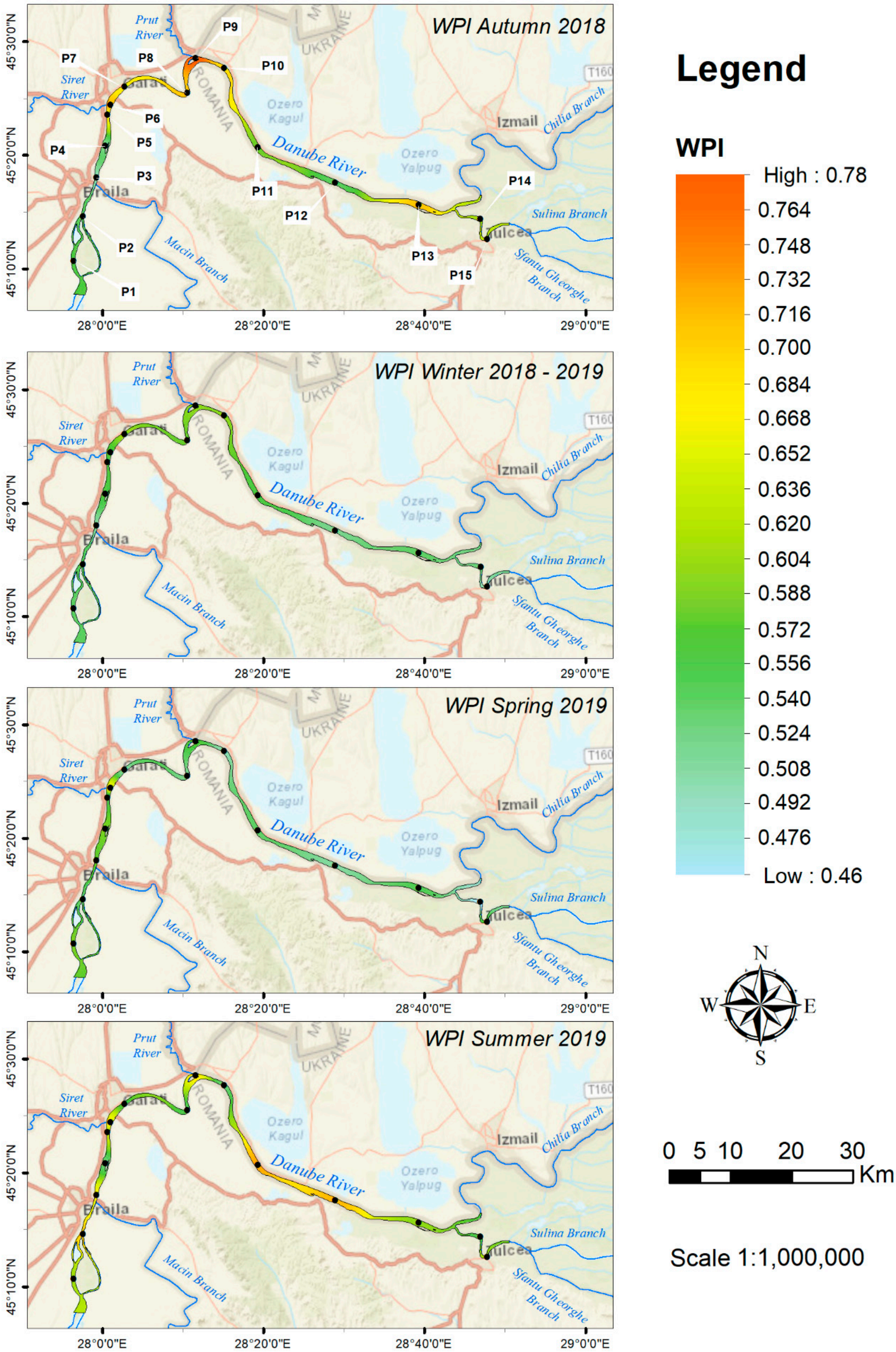

Figure 3. Spatial and temporal distribution of the WPI scores in the Romanian Lower Danube sector recorded during the period autumn 2018–summer 2019.

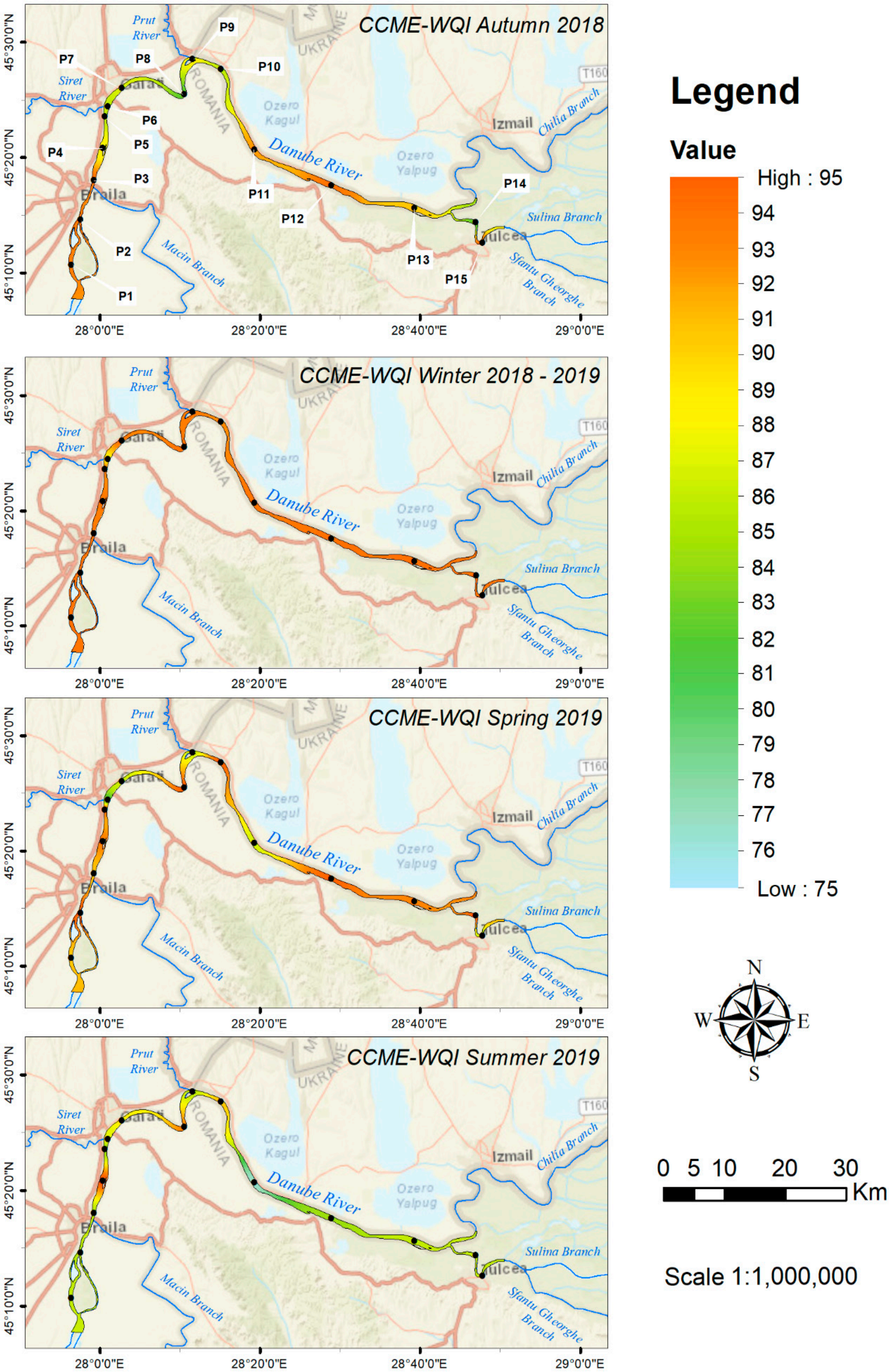

Figure 4. Spatial and temporal variation of the CCME-WQI scores in the Romanian Lower Danube sector recorded during the period autumn 2018–summer 2019.

3.2. A Comparative Approach to the WQI, WPI and CCME-WQI Results

In order to make a comparative approach to the three indices calculated in this study, the weight of each monitored parameter and the seasonal value of the indices for WQI and WPI were represented graphically (Figures 5 and 6). In the case of the CCME-WQI, Table 7 summarizes the values of the three 3 factors which formed the basis of the final results. The percentage weight of each parameter to the final value of the WQI was established based on the value of the $W_i \times q_i$ sub-index in the calculation in equation (1). The results presented in Figure 5 show that Cr-total and N-NO_2^- are the parameters which have the most significant contribution in all the four seasons, although they did not exceed the maximum allowable concentrations (with rare exceptions for N-NO_2^-). These results are explained by the fact that the WQI is generally influenced by those parameters whose maximum allowable concentration is really low (e.g., heavy metals, N-NO_2^-). Actually, the main advantage of using the WQI consists in the fact that the parameters taken into account have a different weight depending on their level of toxicity and on their impact on the aquatic environment. Therefore, this index is appropriate in assessing water quality, especially when determining the level of potentially toxic pollutants in water. The disadvantage of using the WQI, highlighted in the study conducted by Iticescu et al. [49], is that this index has a higher sensitivity to certain parameters (e.g., Cd^{2+}), with the very high values obtained ranking the water analysed in the low-quality class, despite the fact that the experimental values obtained are below the maximum concentration allowed (MCA) by law. Consequently, it is recommended that other indices, which do not limit the integration of some variables, should be tested.

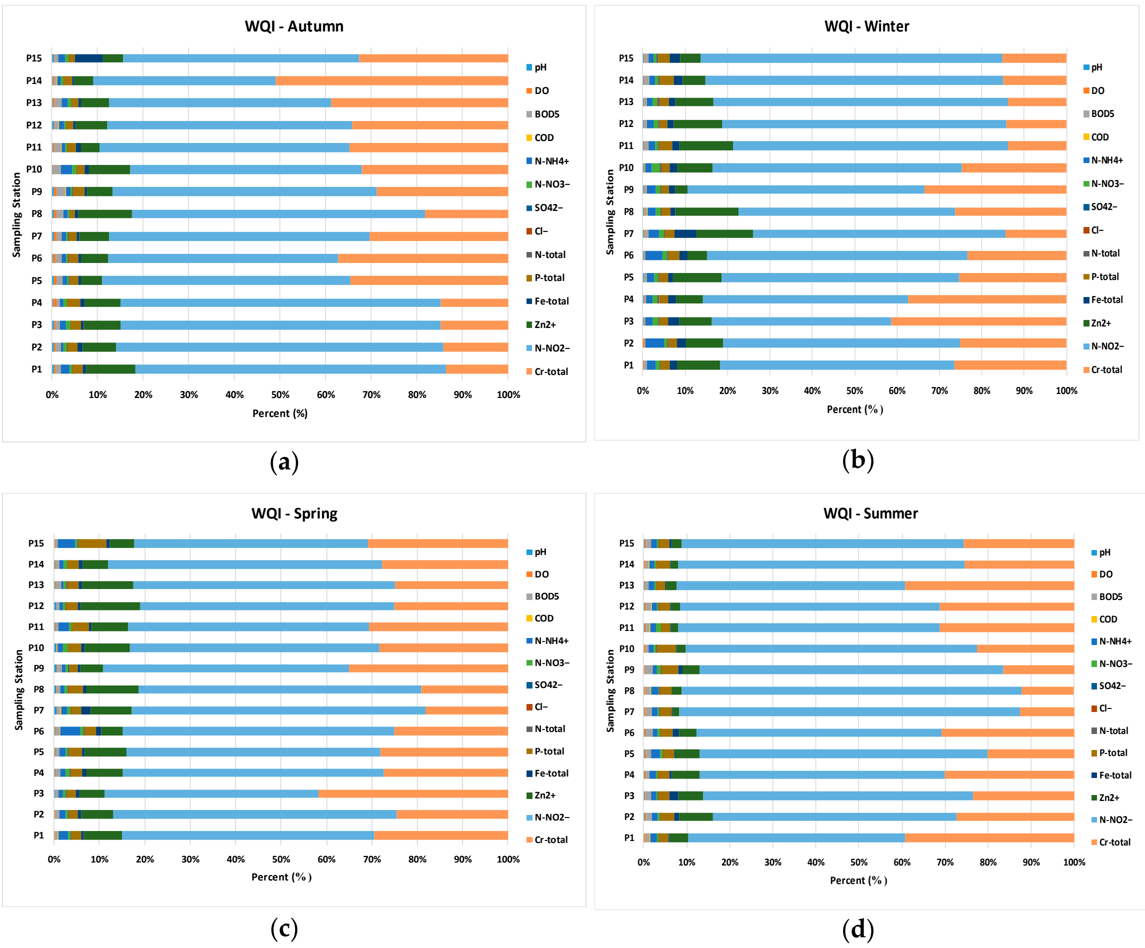

Figure 5. The weight of each parameter to the WQI values recorded in autumn (**a**), winter (**b**), spring (**c**) and summer (**d**).

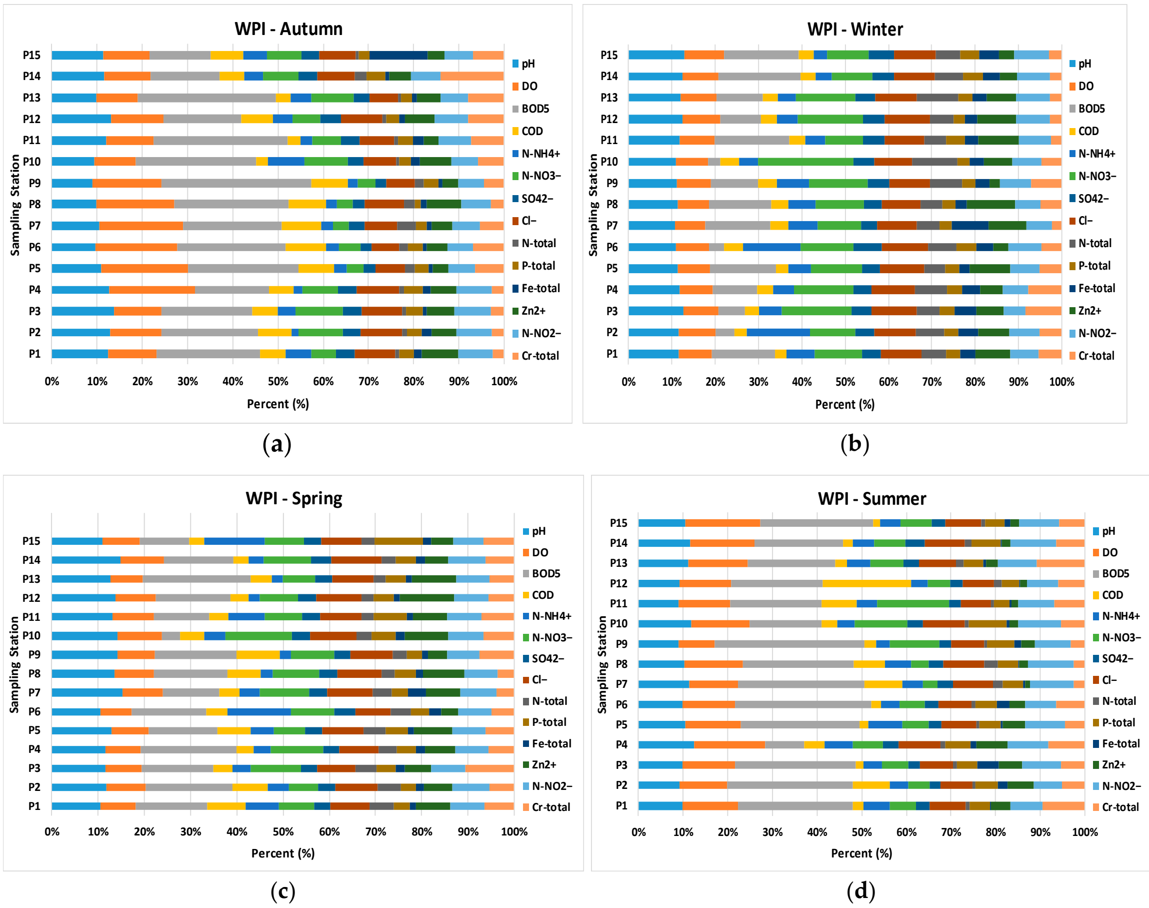

Figure 6. The weight of each parameter to the WPI values recorded in autumn (**a**), winter (**b**), spring (**c**) and summer (**d**).

Figure 6 illustrates the weights of the 14 parameters analysed at the seasonal value of WPI, with these parameters being established according to the result of the A_i/T division (see Equation (2)). According to the graphic representations, the weight of the parameters varies from one season to another. BOD and DO contribute the largest extent to the final results of WPI recorded during summer and autumn. This is due to the fact that the two variables significantly exceeded the maximum allowable concentration in the above-mentioned seasons. The pH parameter also has an important contribution to WPI due to the small range of values in which the experimental results may vary. This is explained by the fact that the maximum allowed pH concentration is a value close to the experimental results. Hence, the value of the A_i/T ratio is higher as compared to the other parameters.

The analysis of the spatial distribution of the weights point to the fact that they differ in the parameters which exceeded the maximum allowed concentration. For example, the contribution of $N\text{-}NH_4^+$ recorded at stations P6 and P15 is much higher than usual, with the recorded values exceeding the MCA during spring. The Water Pollution Index, as opposed to the WQI, does not have a high sensitivity to certain parameters. This means that the WPI calculation formula assigns the same weight to all variables. Consequently, no differentiation may be made between the pollutants according to their ecological impact on the aquatic ecosystem. For example, the effects of a high nitrogen concentration in water are completely different from the effects of the same Cr-total concentration. Therefore, the WPI can be used, especially to determine the level of pollution in the monitored watercourse, with the results showing only the exceedances of the maximum allowed concentrations.

Compared to the WQI and the WPI, the algorithm used for calculating the CCME-WQI does not integrate any sub-indexes. This algorithm is based on the incorporation of 3 factors (F1, F2, F3) which describe the scope, frequency and amplitude of the parameters, all of which were not below the maximum limit allowed by the legislation in force. In order to establish which factor influenced the index scores the most, the values of the 3 factors aggregated in the CCME-WQI seasonal results were compared and summarised (see Table 7). The three factors recorded both seasonal and spatial variations. F1 values fluctuate from 7.14 to 35.71, with the highest value being registered at station P11 in summer. This result significantly influenced the overall value of the index, with the lowest CMME-WQI score (76.54) being obtained at the same time. As explained in Section 3.1, the large number of parameters which did not meet the objective (5 out of 14) was the main cause of the index value obtained. F2 quantifies the number of tests which did not meet the objective. In contrast to the other two indices calculated in this study, the CCME-WQI measures, by means of F2, how many times a parameter exceeded the allowable concentration in a season (frequency). For example, the arithmetic means of monthly tests was used to calculate the seasonal value of the WQI and the WPI, while the CCME-WQI was used to quantify each test individually. The maximum F2 values (i.e., 14.29 and 17.86) are due to the fact that parameters such as BOD, N-NO_3^- and DO recorded values above the allowed limits in a station at least twice in the same season. Thus, this index is suitable for assessing water quality in areas where there are permanent sources of pollution. Another advantage of this index, which was not highlighted in the present paper, is that it allows for the inclusion of a microbiological parameter (faecal coliform) in the calculation formula. Aggregation of physical-chemical, microbiological and biological parameters into a single water quality index is a real challenge which may be a topic of interest for future research.

Based on the aspects presented above, the fact may be observed that the means of aggregating the parameters in the calculation formula represent the main criterion taken into account for determining the usefulness of a water quality index and the individual contribution of the parameters to the final result.

In order to establish the statistical relationship between the three quality indices, the Pearson matrix correlation and scatterplots were applied in this study. Based on the results shown in Table 8, the fact may be observed that significant positive correlations were obtained between the WQI and WPI in spring and autumn. In opposition, significant negative correlations were recorded between CCME-WQI and the other two indices. Negative correlation is due to the fact that the CCME-WQI scores are interpreted differently in comparison with the WPI and WQI scores. For example, a higher value of the CCME-WQI indicates good water quality, while high WPI and WQI values classify water in a lower quality class. Negative correlations were also highlighted in Figure 7f–l, where the relationships between CCME-WQI vs. WQI and CCME-WQI vs. WPI show a downhill pattern.

Table 7. Temporal and spatial variation of F1, F2, F3 and CCME-WQI values.

	F1				F2				F3				CCME-WQI Values			
Season/ Sampling Station	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring	Summer
P1	7.14	7.14	14.29	21.43	7.14	7.14	4.76	10.71	5.41	0.71	6.00	4.90	93.39	94.15	90.64	85.88
P2	7.14	7.14	7.14	21.43	7.14	7.14	2.38	10.71	4.11	0.88	4.93	7.24	93.70	94.15	94.80	85.55
P3	7.14	7.14	14.29	21.43	7.14	7.69	4.76	10.71	2.78	2.33	5.67	6.08	93.95	93.79	90.71	85.73
P4	14.29	7.14	7.14	7.14	14.29	7.69	4.76	3.57	4.04	0.47	7.12	0.64	88.10	93.93	93.56	95.37
P5	21.43	7.14	7.14	21.43	10.71	7.69	2.38	14.29	6.35	1.52	4.93	9.93	85.69	93.88	94.80	84.06
P6	14.29	14.29	28.57	14.29	14.29	14.29	9.52	10.71	13.63	1.12	11.89	12.11	85.93	88.32	81.31	87.54
P7	14.29	7.14	21.43	14.29	14.29	7.14	9.52	10.71	11.15	2.10	3.07	8.26	86.68	94.04	86.35	88.64
P8	14.29	7.14	7.14	7.14	10.71	7.14	4.76	7.14	4.87	1.13	2.10	6.10	89.31	94.13	94.90	93.19
P9	14.29	7.14	21.43	14.29	14.29	7.69	7.14	14.29	19.05	1.87	4.42	14.04	83.97	93.84	86.71	85.80
P10	7.14	7.14	7.14	14.29	7.14	7.69	4.76	7.14	10.26	6.25	2.02	3.56	91.69	92.95	94.91	90.55
P11	7.14	7.14	21.43	35.71	7.14	7.14	7.14	17.86	9.09	2.37	3.98	7.25	92.15	94.01	86.76	76.57
P12	7.14	7.14	7.14	21.43	7.14	7.14	4.76	10.71	2.10	0.71	2.46	12.31	94.04	94.15	94.84	84.45
P13	7.14	7.14	7.14	21.43	7.14	7.14	7.14	10.71	12.83	0.24	5.32	3.30	90.57	94.17	93.41	86.04
P14	14.29	7.14	7.14	21.43	14.29	7.14	4.76	10.71	23.91	2.78	1.22	2.40	81.93	93.95	94.99	86.10
P15	7.14	7.14	21.43	21.43	7.14	7.14	7.14	10.71	1.41	1.55	6.73	5.71	94.11	94.10	86.39	85.78

Table 8. Correlation matrix for the WQI, WPI and CCME-WQI results.

Variable	WQI-Autumn	WPI-Autumn	CWQI-Autumn	CWQI-Winter	WPI-Winter	CWQI-Winter	WQI-Spring	WPI-Spring	CWQI-Spring	WQI-Summer	WPI-Summer	CWQI-Summer
WQI-Autumn	1.00											
WPI-Autumn	0.50	1.00										
CWQI-Autumn	−0.46	−0.57	1.00									
WQI-Winter	−0.28	0.41	−0.21	1.00								
WPI-Winter	−0.04	0.59	−0.54	0.57	1.00							
CWQI-Winter	−0.18	−0.32	0.26	−0.42	−0.18	1.00						
WQI-Spring	−0.44	−0.32	−0.56	0.23	−0.34	−0.33	1.00					
WPI-Spring	−0.40	0.02	0.28	0.46	−0.10	−0.63	0.72	1.00				
CWQI-Spring	0.01	−0.29	−0.14	−0.15	−0.15	0.57	−0.41	−0.62	1.00			
WQI-Summer	0.11	−0.32	0.45	−0.20	−0.60	0.03	0.52	0.26	−0.03	1.00		
WPI-Summer	−0.18	−0.18	0.60	0.15	−0.28	−0.14	0.45	0.28	−0.33	0.35	1.00	
CWQI-Summer	−0.19	0.14	−0.47	0.24	0.30	−0.09	−0.26	0.08	0.26	−0.52	−0.76	1.00

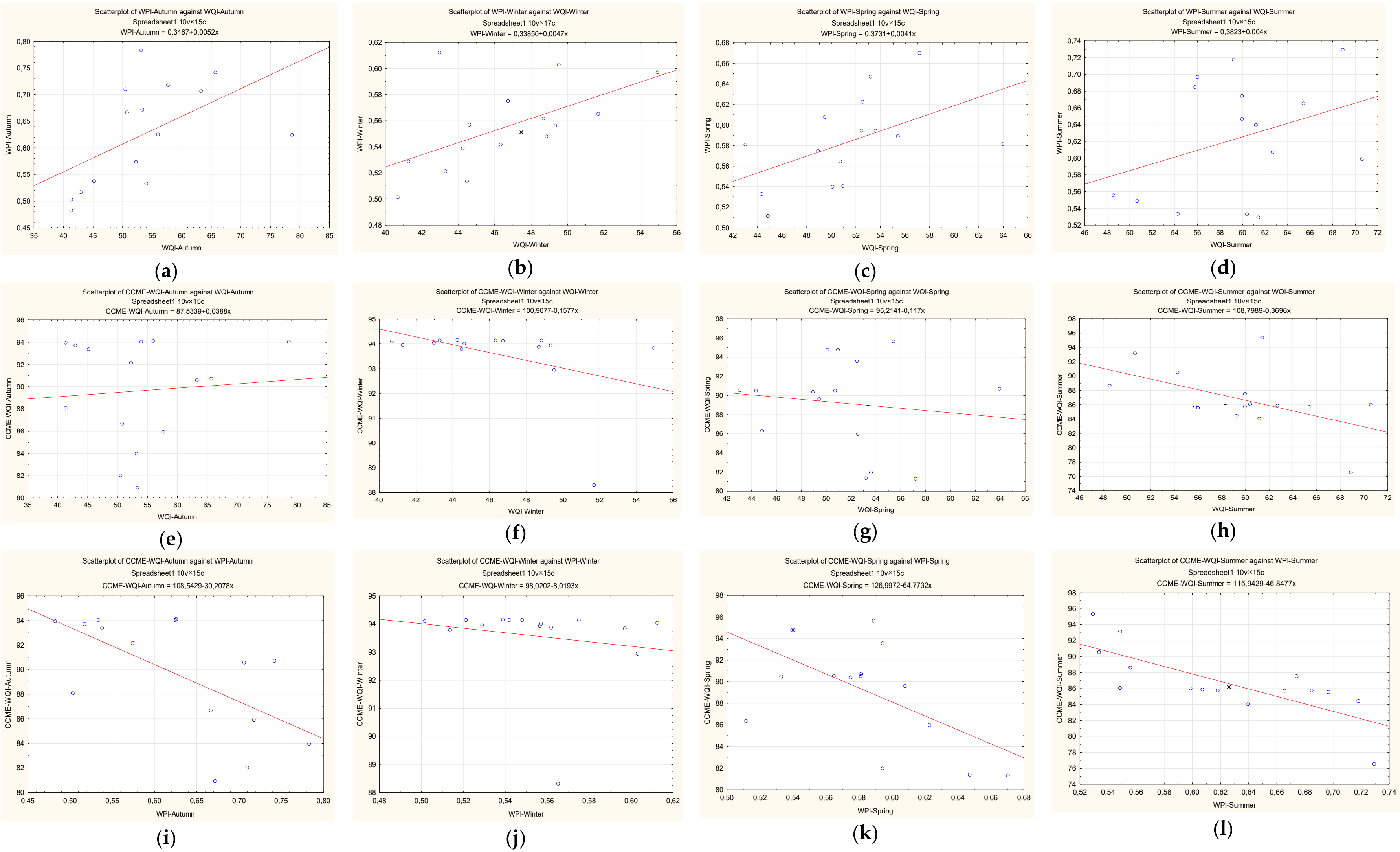

Figure 7. Scatterplots showing the relationships between WPI vs. WQI (**a**,**b**,**c**,**d**), CCME-WQI vs. WQI (**e**,**f**,**g**,**h**) and CCME-WQI vs. WPI (**i**,**j**,**k**,**l**).

4. Conclusions

The Water Quality Index, Water Pollution Index and Canadian Council of Ministers of the Environment Water Quality Index were applied and compared in this study in order to assess water quality in the Lower Danube.

The results of the spatial assessment provided information on the existence of pollution sources. The water quality in the study area was found to be lower in the sampling stations located near the areas where agricultural and industrial activities take place. Moreover, the spatial distribution of indices values highlighted the fact that the Danube water quality is influenced by its major tributaries (Rivers Siret and Prut) through the intake of pollutants they transport. In terms of water quality seasonal variation, a lower quality was observed during summer and autumn, with this being due to flow variations and to the parameters which vary according to temperature.

Based on the results obtained, the Danube water quality was evaluated differently in relation to the three indices. According to the WQI index, 53% of sampling stations were classified in quality class III, while 47% were classified in quality class II. Compared to the WQI, the results obtained by using the WPI ranked water strictly in quality class II ("pure quality"). Approximately similar to the WPI, the CCME-WQI classified the water analysed in 98% of all sampling stations in quality class II. The differences between the three indices were due to the weight (contribution) of each aggregate parameter in the calculation formula. Thus, the WQI will be applied in the evaluation of water quality, especially when determining the level of potentially toxic pollutants in water. The CCME-WQI will be applied when assessing water quality in areas where there are permanent sources of pollution. As regards the WPI, its low sensitivity to certain parameters makes it appropriate for a general characterisation of watercourses. Therefore, it is necessary that a suitable index should be chosen and applied depending on the ecosystem complexity, on the type(s) of pollution sources and on the purpose of the monitoring activity.

The general conclusion is that, among the three indices, WQI seems to be the most suitable in assessing water quality in the study area due to the various types of existing pollution sources.

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