LONG-TERM WATER QUALITY TRENDS IN THE LOWER DANUBE (1996-2017)

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The paper presents the trends of the main water quality parameters during the period 1996-2017 in the Lower Danube and in 6 important tributaries from Romania. 14 physical-chemical parameters were analysed using statistical methods: t test, Spearman rank correlation coefficient, as well as multivariate methods (principal component analysis and factor analysis). In most cases, water quality has improved during the analysed period, but there were some cases in which certain parameters have deteriorated. Principal Component Analysis on annual averaged values has revealed the largest variations for ammonium and total phosphorus.

Keywords: Danube water quality, Student t test, Spearman rank correlation, principal component analysis, factor analysis

1. Introduction

The Danube is the second longest river in Europe, and it plays an important role for riparian countries as a source of drinking and irrigation water, waterway for navigation, energy source in hydro-power stations, having at the same time cultural and recreational value. It passes many regions with different topography and climate and it hosts a complex aquatic ecosystem, which is influenced by pollution generated by human activities, such as agriculture, land use, mining, urban agglomerations [1].

The International Commission for the Protection of the Danube River (ICPDR) coordinates the efforts of the participating countries to control pollution and improve water quality in the Danube Basin. Monitoring data are reported within the TransNational Monitoring Network (TNMN) and are stored in the water quality database of the ICPDR [2].

Long term water quality trends in the Danube have been studied by Jaruskova and Liska (2011) for the period 1996-2005 [3] and by Hamchevici and Udrea (2015) for the period 2001-2009 [4]. The TNMN publishes monitoring data in its Yearbooks, including comparisons for up to 11 years [5].

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Danube water quality in the Romanian sector has been studied, for shorter periods of time, by several authors, including Ismail and Robescu, who focused on the section between the Iron Gates I and Drobeta Turnu Severin [6, 7], Iticescu et al., who studied the area of Galati [8-10], Radu et al., who studied the section between km 375 and 175 [11-13].

This paper examines monitoring data for a period of 22 years at 9 stations along the Lower Danube and 6 on its main tributaries from Romania. It includes comparisons between the Danube and the tributaries, and it analyses their impact on its water quality. Statistical methods (t test, Spearman rank correlation, PCA and factor analysis) were used to assess the evolution in time of 14 parameters at 15 monitoring stations, to differentiate pollution levels along the Danube, identify correlations between parameters and differences between locations.

2. Data and Methods

The study area consists of 15 monitoring stations of the TNMN network, 9 located on the Danube, between km 1071 and km 0, and 6 on the tributaries Jiu, Olt, Argeș, Ialomița, Siret and Prut. The locations of the monitoring stations are presented in Fig. 1 [14], and more details are included in Table 1 [5].

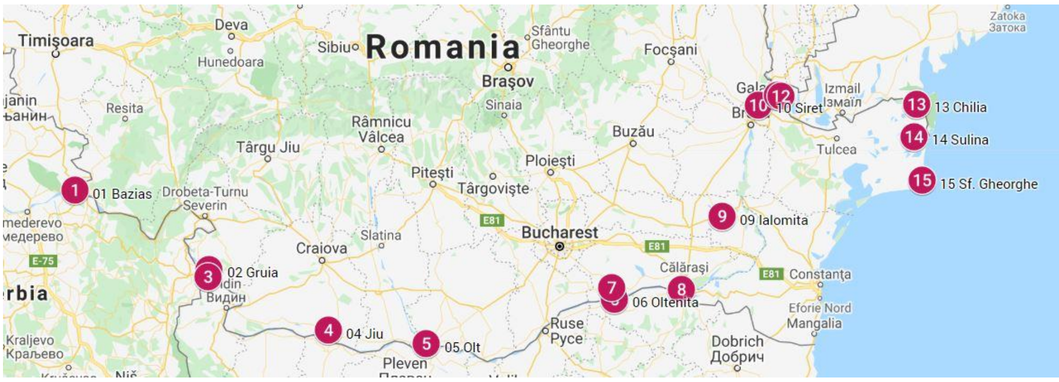

Fig. 1. Positioning on the map of the studied monitoring stations [14]

Monitoring data for the period 1996-2017 were retrieved from the ICPDR Water Quality Database [2], with the permission to use them for scientific purposes.

Table 1

Location of the studied monitoring stations [5]

Location name	TNMN location code	Longitude	Latitude	River	Position along the river (km)	Catchment area (km^2)
01 Bazias	RO1	21.38397	44.81610	Danube	1071.0	570896.00
02 Gruia	RO18	22.68355	44.27018	Danube	851.0	577085.00

03 Pristol	RO2	22.67613	44.21418	Danube	834.0	580100.00
04 Jiu	RO19	23.84549	43.84188	Jiu	9.0	10046.00
05 Olt	RO20	24.79680	43.74400	Olt	3.0	24050.00
06 Oltenita	RO3	26.61905	44.05605	Danube	432.0	676150.00
07 Arges	RO9	26.59900	44.14500	Argeș	0.0	12550.00
08 Chiciu	RO4	27.26771	44.12757	Danube	375.0	698600.00
09 Ialomita	RO21	27.66468	44.63477	Ialomița	24.0	10309.00
10 Siret	RO10	28.00940	45.41474	Siret	0.0	42890.00
11 Prut	RO11	28.20300	45.46900	Prut	0.0	27480.00
12 Reni	RO5	28.23190	45.46324	Danube	132.0	805700.00
13 Chilia	RO6	29.55336	45.40635	Danube	18.0	817000.00
14 Sulina	RO7	29.52966	45.18338	Danube	0.0	817000.00
15 Sf Gheorghe	RO8	29.60945	44.88462	Danube	0.0	817000.00

For some locations, sampling is done on the left bank (L), in the middle (M) and on the right bank (R). In this work only data from the middle sampling points (M) were used, because they were available for all stations.

Samples were taken, processed and analysed according to standard methods by the National Administration “Romanian Waters” (ANAR), who is the responsible authority for water quality monitoring in Romania, and reported to the TNMN. Most parameters are analysed monthly, so the database includes hundreds of observations for each parameter and location from the period 1996-2017. Some locations were included in the monitoring network in 2006 (Gruia, Jiu, Olt, Ialomița), so in these cases data are only available for 11 years.

The following statistical methods were used for data analysis:

- Student t test is used to compare the means of two datasets and decide if there are significant differences between them. For each parameter, the values from Bazias, the first monitoring station in Romania, were compared to the values from all the other sampling points, in order to see if there are significant increases or decreases along the Danube, as it flows towards the Black Sea;
- Spearman Rank Correlation test is used to find out if there is a monotonic correlation between two variables (linear or not), which uses ranks instead of variables values. Spearman test is preferred in the case of datasets that do not have a normal distribution, because it is not sensitive to outliers. In this work Spearman rank correlation was used to identify time trends in the values of the analysed parameters.
- Principal Component Analysis (PCA) is applied for large datasets with many variables that may be correlated, in order to reduce the number of dimensions and identify relationships between variables. The principal components are linear combinations of the original variables, and each component explains a proportion of the variation of the dataset. The first component explains the

largest part of the variance, the second explains the second largest part, and so on. In this way, some components may be discarded without losing much of the information contained in the dataset. In this study, PCA was used to identify which parameters play the most important roles in the variation of the dataset and to find correlations between parameters [15].

- Factor analysis takes into account at the same time the numerical values of all the parameters and the location where the sample was taken. It was used to analyse the differences between sampling locations and highlight the locations with a different profile than in the others.

Trend analysis was applied for daily average flow, water temperature (WT), pH, dissolved oxygen (DO), biological oxygen demand (BOD5), chemical oxygen demand (CCO-Mn, CCO-Cr), ammonium (N-NH4), nitrites (N-NO2), nitrates (N-NO3), total nitrogen (TN), orthophosphates (P-PO4) and total phosphorus (TP).

PCA and factor analysis were applied for mean annual values of pH, DO, BOD5, CCO-Cr, N-NH4, N-NO2, N-NO3, P-PO4, TP, chlorides, sulphates and suspended solids.

Data were downloaded in an Excel table and further processed using Microsoft Excel and R statistical software version 3.6.2.

3. Results and Discussion

3.1. Comparison between parameter values at Bazias and the other stations

The results of the t test, comparing the values from each station to the values from Bazias, are presented in Table 2.

For water temperature there were no significant differences between the studied locations and the values from Bazias. Water temperature is closely correlated with air temperature, and influences gas solubility in water, which decreases as temperature rises.

pH was lower in the Olt, Arges and Ialomita rivers compared to Bazias and higher at Chiciu, Reni and in the Siret and Prut rivers. However, there was no significant difference between the pH values at Bazias and the ones at Danube mouths, where it flows into the Black Sea (Chilia, Sulina, Sf. Gheorghe).

Dissolved oxygen values were lower in the Arges River compared to Bazias, and higher at Jiu, Oltenita, Chiciu and Reni. Oxygen saturation was lower in the Arges, Siret and Prut rivers and higher at Jiu, Oltenita and Reni. There were no significant differences between Bazias and Danube mouths for DO and OS. Oxygen is important for aquatic life, but algal growth in excess may lead to oxygen depletion.

Table 2

Comparison between monitoring values calculated by Student t statistic at Bazias and the other locations for the period 1996-2017

Comparison between station 01 Bazias and station	Parameter												
	WT	pH	DO	OS	BOD5	CCO-Mn	CCO-Cr	N-NH4	N-NO2	N-NO3	TN	P-PO4	TP
02 Gruia*	ns	ns	ns	ns	<	<	>	<	<	ns	ns	ns	>
03 Pristol	ns	ns	ns	ns	ns	ns	ns	<	ns	ns	ns	ns	ns
04 Jiu*	ns	ns	>	>	>	>	>	<	<	>	>	<	<
05 Olt*	ns	<	ns	ns	<	<	>	ns	<	<	ns	<	<
06 Oltenita	ns	ns	>	>	>	>	>	<	ns	>	ns	<	ns
07 Arges	ns	<	<	<	>	>	>	>	>	>	>	>	>
08 Chiciu	ns	>	>	ns	<	>	>	>	ns	>	>	<	ns
09 Ialomita*	ns	<	ns	ns	>	>	>	>	>	>	>	>	>
10 Siret	ns	>	ns	<	>	>	>	>	>	>	>	<	ns
11 Prut	ns	>	ns	<	>	>	>	>	ns	>	>	<	<
12 Reni	ns	>	>	>	<	>	>	ns	ns	>	>	<	ns
13 Chilia	ns	ns	ns	ns	<	>	>	>	ns	>	>	<	<
14 Sulina	ns	ns	ns	ns	ns	>	>	>	ns	>	>	<	<
15 Sf Gheorghe	ns	ns	ns	ns	<	>	>	ns	ns	>	>	<	<

*Monitoring started in 2007; ns = not significant; < values were significantly lower than at Bazias; > values were significantly higher than at Bazias; - better water quality; - worse water quality

BOD5 values were lower at Gruia, Olt, Chiciu, Reni, Chilia and Sf. Gheorghe, and higher at Jiu, Oltenita, Arges, Siret and Prut. BOD5 is an indicator for the presence of biodegradable organic matter in water.

CCO-Mn values were lower at Gruia and Olt and higher at all the other stations except Pristol. CCO-Cr was higher at all stations compared to Bazias, except for Pristol. These two parameters indicate that organic pollution increased between Bazias and the Black Sea. Potassium permanganate is a weak oxidizing agent, and CCO-Mn has similar values with BOD5, but it can be determined much faster, in a few hours, instead of 5 days. CCO-Cr is a strong oxidizing agent, which can break down organic matter that is not easily biodegradable, like lignin and cellulose.

Ammonium levels were lower at Guia, Pristol, Jiu and Oltenita, and higher at Arges, Chiciu, Ialomita, Siret, Prut, Chilia and Sulina. Ammonium is often an indication of insufficiently treated municipal wastewater discharged into surface water bodies.

Nitrites levels were lower at Gruia, Jiu and Olt, And higher at Arges, Ialomita and Siret. Nitrites are unstable compounds that oxidize to nitrates in aerobic conditions.

Nitrates levels were lower in the Olt River, and higher at all the other stations except for Gruia and Pristol. Nitrates may enter water bodies via runoff from agricultural land where fertilizers are applied.

Total nitrogen levels were higher at all stations downstream compared to Bazias, except for Gruia, Pristol, Olt and Oltenita, where the test was not significant.

Orthophosphates levels were higher compared to Bazias in the Arges and Ialomita rivers and lower at all the other stations, except Gruia and Pristol. Total phosphorus levels were higher at Gruia, Arges and Ialomita, and lower at Jiu, Olt Prut, Chilia, Sulina and Sf. Gheorghe. Phosphorus parameters were significantly lower during the studied period downstream compared to Bazias. Phosphates used to enter water bodies over municipal wastewater, because many detergents were based on them. The use of phosphates in detergents was banned in the EU since 2013.

3.2. Trend analysis for the period 1996-2017

Spearman rank correlation coefficients were calculated for each parameter at each location between determined values and sampling date. Positive correlations indicate that values have increased in the studied period, while negative coefficients indicate that values have decreased between 1996-2017. In order to assess if trends were significant from statistical point of view, p values (probability that test is significant) were calculated at 0.95 confidence level, and trends were considered significant if $p < 0.05$.

The results of the trend analysis are presented in Table 3.

Daily average flow has increased only in Arges River, while at all the other stations it has decreased, except for Sf. Gherghe where there is no significant trend.

Water temperature has increased only at Chiciu and Prut, while at the other stations there was no significant trend.

pH has increased at all stations except Arges, where it decreased, while at Ialomita, Siret and Prut the trend was not significant.

DO trend was not significant at 7 stations and increased at 8 stations, particularly in the final section of the Danube. OS has decreased at Jiu, Oltenita and Arges, and was not significant in the Olt and Ialomita rivers, while at all the other stations it has increased, so water quality has improved.

Table 3

Trend analysis of water quality parameters determined by Spearman rank correlation coefficients at selected locations for the period 1996-2017

Location	Parameter													
	Flow	WT	pH	DO	OS	BOD5	CCO-Mn	CCO-Cr	N-NH4	N-NO2	N-NO3	TN	P-PO4	TP
01 Bazias	↓	ns	↑	ns	↑	↓	↓	↑	↓	↓	↓	ns	↓	↓
02 Gruia*	-	ns	↑	ns	↑	ns	↓	↑	↓	ns	↓	ns	↓	↓
03 Pristol	↓	ns	↑	↑	↑	↓	↓	↑	↓	↓	↓	ns	↓	↓
04 Jiu*	-	ns	↑	ns	↓	↑	↑	↑	↓	↓	↓	ns	ns	ns
05 Olt*	-	ns	↑	ns	ns	ns	ns	↓	↓	↓	↑	↓	ns	↓
06 Oltenita	↓	ns	↑	ns	↓	↑	↑	↑	↓	↓	↓	↓	↓	↓
07 Arges	↑	ns	↓	ns	↓	↓	↓	↑	ns	↑	↓	↓	↑	ns
08 Chiciu	↓	↑	↑	↑	↑	↓	↓	ns	↓	↓	↓	↓	↑	ns
09 Ialomita*	-	ns	ns	ns	ns	ns	ns	ns	ns	↓	ns	↓	↑	ns
10 Siret	↓	ns	ns	↑	↑	↓	↓	↓	↓	↓	↓	↓	↓	↓
11 Prut	↓	↑	ns	↑	↑	↓	↓	↓	↓	↓	↓	↓	ns	ns
12 Reni	↓	ns	↑	↑	↑	↓	↓	↓	↓	↓	↓	↓	↑	ns
13 Chilia	↓	ns	↑	↑	↑	↓	↓	↓	↓	↓	↓	↓	ns	ns
14 Sulina	↓	ns	↑	↑	↑	↓	↓	↓	↓	↓	↓	↓	ns	ns
15 Sf Gheorghe	ns	ns	↑	↑	↑	↓	↓	↓	↓	↓	↓	↓	ns	ns

*Monitoring started in 2007; - no data available; ns = not significant; ↑ values have increased during the studied period; ↓ values have decreased during the studied period; - water quality has improved; - water quality has deteriorated.

BOD5 has decreased at 10 stations increased at Jiu and Oltenita and was not significant at 3 stations (Gruia, Olt, Ialomița). A study on the period 1996-2005 also found decreasing trends at Pristol, Oltenița, Chiciu and Reni [3].

CCO-Mn decreased at 11 stations, increased at Jiu and Oltenita and was not significant at two stations. These two parameters are related, as they reflect the amount of biodegradable matter present in the water, and the analysis shows that they had similar trends during the studied period.

CCO-Cr has increased at 6 stations (among the first 7) and decreased at 7 stations, particularly in the last section, before flowing into the Black Sea. At Chiciu and Ialomita no significant trend was detected. A previous study found decreasing trend at Pristol, increase at Chiciu and Reni and no significant trend at Oltenita [3].

Ammonium has decreased at all stations except Arges and Ialomita, which may be an indication of improved wastewater treatment capacity on the other tributaries. Previous studies (1996-2009) only found decreasing trends at some of the stations, so the situation has improved after 2009 [3, 4].

Nitrites have increased on the Arges River, had no significant trend at Gruia, and decreased at all the other stations. Other studies found similar results for periods until 2009 [4].

Nitrates have increased in the Olt river, had no significant trend in Ialomita River and decreased at all the other stations. Studies for shorter periods found slightly different results, so the situation appears to have improved after 2009 [3] [4].

Total nitrogen had no significant trends at Bazias, Gruia, Pristol and Jiu, and decreased at all the other stations. This a positive trend, that shows that efforts to reduce nitrogen release to water bodies were mostly successful. Decreasing amounts of nitrogen discharged into the Black Sea were confirmed for the period 1996-2002 at Sulina [16], but for the period 2001-2009 the test results were not significant [4].

Orthophosphates have decreased at Bazias, Gruia, Pristol Oltenita and Siret, increased at Arges, Chiciu, Ialomita and Reni, and had no significant trend at the other locations. Previous studies found increasing trends at Pristol, and no significant trend at Chiciu and Reni, so phosphorus discharges affecting these stations could have intensified after 2009 [3, 4].

Total phosphorus has decreased at 6 stations and had no significant trend at 9 stations. Other studies on shorter periods found increasing trends at Bazias, Pristol and Reni, so the situation could have improved after 2009 [3, 4].

3.3. Principal Component Analysis (PCA)

PCA was applied on annual means of 12 parameters in order to identify which parameters had the highest variation during the studied period and to highlight existing correlations between parameters. The analysed parameters were pH, DO, BOD5, CCO-Cr, N-NH4, N-NO2, N-NO3, P-PO4, TP, chlorides, sulphates and suspended solids. Data were centered and scaled before analysis to remove the effect of measuring units and orders of magnitude. PCA results are presented in Fig. 2 and Table 4. The first component PC1 (Dim1) explains 36.77% of the variation of the dataset, and is dominated by ammonium, TP, sulphates and BOD5. PC2 (Dim2) explains 17.10% of the variation and is dominated by CCO-Cr, P-PO4 and chlorides. The first 8 components explain over 90% of the variation of the dataset, so the number of variables could be reduced from 12 to 8 without losing important information.

Table 4

PCA results on mean annual values at selected stations for the period 1996-2017

Parameter	**PC1**	**PC2**	**PC3**	**PC4**	**PC5**	**PC6**
pH	-0.1797	0.1594	-0.6907	0.0343	0.2384	-0.5182
Dissolved Oxygen	-0.2201	0.2770	0.0471	0.2971	0.6797	0.2678
BOD5	0.3263	-0.0900	-0.2744	0.0277	-0.3327	-0.0176
CCO-Cr	0.2049	0.4605	0.1184	0.2549	-0.1749	-0.4641
N-NH4	0.3833	-0.1944	0.0774	-0.1430	0.1529	-0.0274
N-NO2	0.2667	0.0062	0.3735	-0.3901	0.4055	-0.4655
N-NO4	0.2728	0.1384	-0.2952	-0.5520	0.1294	0.1641
P-PO4	0.3046	-0.4171	-0.0649	0.3704	0.1666	0.0135
Total P	0.3422	-0.2990	-0.0692	0.3913	0.2253	-0.1201
Suspended solids	0.2928	0.3770	-0.0926	0.2452	-0.1687	0.1010
Chlorides	0.2717	0.4210	0.2855	0.1026	0.0031	0.1154
Sulphates	0.3300	0.1951	-0.3158	-0.0809	0.1684	0.4020
Standard deviation	2.1005	1.4324	1.0790	1.0084	0.9105	0.7600
Explained variance (%)	36.77	17.10	9.70	8.47	6.91	4.81
Cumulative explained variance (%)	36.77	53.87	63.57	72.04	78.95	83.76
Parameter	**PC7**	**PC8**	**PC9**	**PC10**	**PC11**	**PC12**
pH	0.0366	-0.1341	0.2183	0.2580	0.0496	0.0529
Dissolved Oxygen	0.3585	0.2663	-0.1816	-0.0636	-0.1194	-0.0018
BOD5	0.8022	0.0668	-0.1457	-0.1385	0.0706	-0.0697
CCO-Cr	-0.1584	0.3816	0.0724	-0.4593	-0.1572	0.0971
N-NH4	0.0773	0.2587	0.3925	0.3445	-0.6447	-0.0591
N-NO2	0.1596	-0.3786	-0.2661	-0.0943	0.0526	0.0683
N-NO4	-0.2219	0.5244	-0.2470	0.0473	0.2859	0.0422
P-PO4	-0.0988	0.0518	-0.0177	-0.0071	0.1843	0.7191
Total P	-0.2252	0.0454	-0.0954	-0.0342	0.2337	-0.6729
Suspended solids	-0.1576	-0.2561	-0.5480	0.4432	-0.2730	0.0548
Chlorides	0.1420	-0.0533	0.4717	0.3612	0.5173	0.0052
Sulphates	-0.1236	-0.4501	0.2733	-0.4913	-0.1510	-0.0040
Standard deviation	0.7006	0.6715	0.6127	0.5363	0.4934	0.3165
Explained variance (%)	4.09	3.76	3.13	2.40	2.03	0.84
Cumulative explained variance (%)	87.85	91.61	94.74	97.14	99.17	100.00

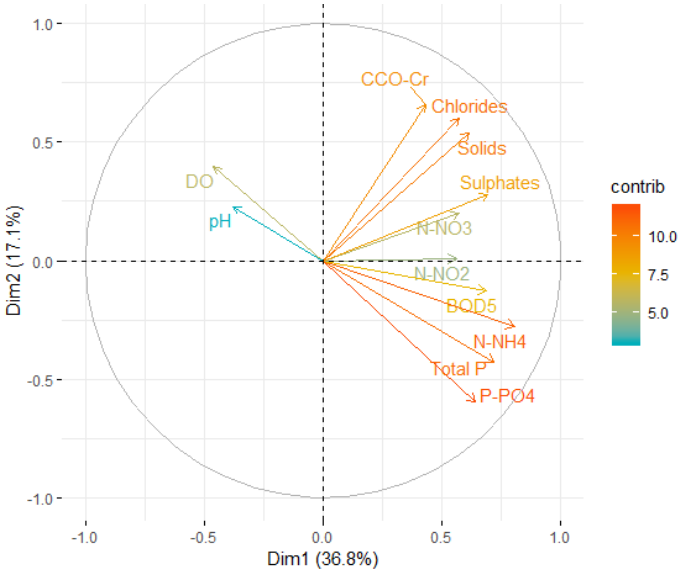

Fig. 2. Graphical representation of the first two principal components (Dim1 and Dim2) for annual mean values at selected monitoring stations for the period 1996-2017

In Fig. 2, arrows represent parameters, and the length of the arrow is proportional with PC1 and PC2 coefficients, so the longer the arrow, the higher the variation of the parameter. Angles smaller than 90º between arrows indicate positive correlations (the smaller the angle, the higher the correlation), angles larger than 90º indicate negative correlations, and right angles indicate no correlation.

The results indicate positive correlations between pH and DO, N-NO2 and BOD5, chlorides and suspended solids. Negative correlations can be noticed between DO and P-PO4, pH and TP. The parameters with the smallest variation are pH, DO, N-NO2 and N-NO3.

3.4. Factor analysis

Factor analysis can highlight at the same time the influence of the parameters, as well as the location where the sample was taken. The results of the factor analysis are shown in Fig. 3, where locations are represented by black triangles.

Fig. 3 highlights that pollution levels in Ialomita and Arges tributaries are much higher than in the Danube. While Ialomita is more influenced by organic pollution (CCO-Cr), chlorides, sulphates, the Arges River is more influenced by ammonium and phosphorus, a clear indication of pollution with insufficiently treated municipal wastewater. In this case, pollution originates from Bucharest wastewater treatment plant, which discharges into Dambovita River, an Arges tributary, as was demonstrated by Ionescu et al. [17].

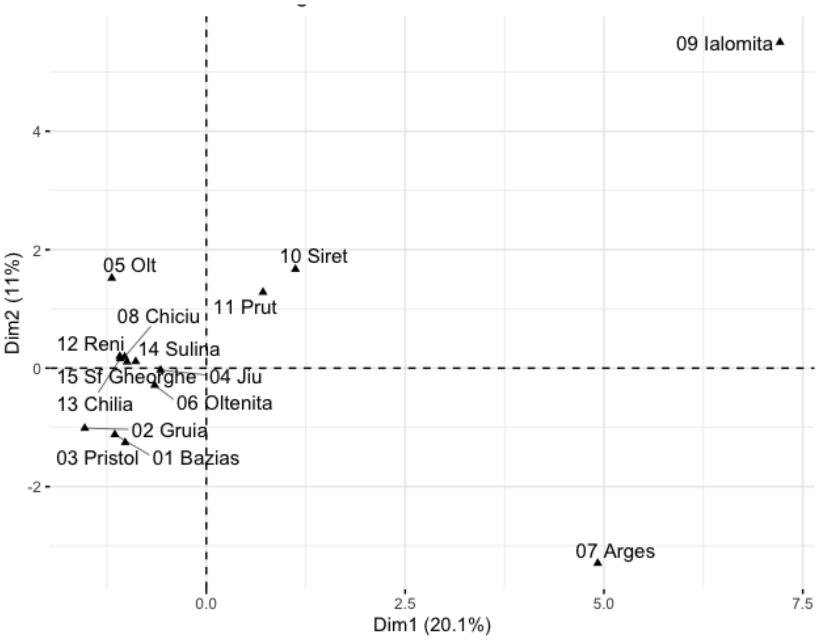

Fig. 3. Factor analysis of annual means and locations for the period 1996-2017

The annual mean values of the first three stations, Bazias, Gruia and Pristol, are very close to those of the last three stations, Chilia, Sulina and Sf. Gheorghe, as was shown also by the t tests.

4. Conclusions

The study has shown that, by comparing parameter values at Bazias, where the Danube enters Romanian territory, with values at the Danube mouths (Chilia Sulina, Sf. Gheorghe), for the period 1996-2017, in some cases water quality improves (BOD5, P-PO4, TP), in others it remains the same (pH, DO, OS, N-NO2), but in others pollution increases (CCO-Mn, CCO-Cr, N-NH4, N-NO3, TN).

Trend analysis reveals that pollution has decreased significantly during the study period for most parameters, particularly in the last section of the Danube, before it flows into the Black Sea.

PCA shows that highest variations were for ammonium, TP, sulphates and BOD5. Positive correlations were found between pH and DO, N-NO2 and BOD5, chlorides and suspended solids. Negative correlations were found between DO and P-PO4, pH and TP.

Factor analysis shows that the high variations originate mainly from the pollution of the Arges River, which receives insufficiently treated municipal wastewater. Pollution levels are also much higher in Ialomita River than in the Danube, but in this case organic compounds, chlorides and suspended solids dominate. Although pollution levels have decreased, efforts are still needed to improve wastewater treatment in large human agglomerations.

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