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IDENTIFICATION OF POLLUTION SOURCES IN THE NAREW RIVER CATCHMENT USING MULTIVARIATE STATISTICAL METHODS

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Abstract:
The aim of the statistical analyses carried out was to identify similarities and to point out differences between the various tributaries of the Narew River, to identify the factors and processes responsible for the transformations occurring in the aquatic environment and finally, to identify the main sources of pollution in the river catchment. For the purposes of statistical analysis, the results of studies conducted as part of diagnostic monitoring by the General Inspectorate for Environmental Protection in 2017–2018 were used. The studies included 8 measurement points located directly on the Narew River and 17 points located on its selected left and right tributaries. Analysis of the collected results indicates that the chemical condition of the water in the Narew catchment is assessed as being poor. This observation may be due to the fact that the Narew catchment is mainly used for agricultural purposes and, in addition, there is a relatively large number of potential anthropogenic sources. As part of the analysis, two potential sources of pollution affecting water quality in the Narew catchment were identified, which include surface run-off and treated wastewater inflow.

Key words: Narew River catchment, pollution sources identification, cluster analysis.

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INTRODUCTION

The quality of flowing surface water is variable over time and depends on a number of factors that are difficult to identify (Ahmadmoazzam *et al.*, 2021). The mechanisms and kinetics of transformations that constituents and pollutants undergo in surface waters consist of processes associated with the deposition of substances in the environment, including among others sorption, desorption and bioaccumulation (Apostolovic *et al.*, 2020; Souza *et al.*, 2020). In addition, processes related to biological transformations also occur in the aquatic environment, among which biodegradation under aerobic and anaerobic conditions is distinguished (Payandi-Rolland *et al.*, 2020; Wang *et al.*, 2021). It should be emphasized that the mentioned processes occur with varied intensity which, mainly in surface watercourses, depends on a seasonal variation related to temperature and, consequently, to the dominant form of a river catchment usage (Xiaohui *et al.*, 2021). In addition to naturally occurring phenomena in a given area, the quality of river water is influenced by anthropogenic factors related to the agricultural use of the land adjacent to river channels or the direct introduction of pollutant streams into water (Tao *et al.*, 2021; Zhang *et al.*, 2021a). Another factor influencing the pollution identification is a size of the river catchment under study (Pu *et al.*, 2021). As a rule, the identification of migration routes of selected pollutants is less complex in the case of smaller catchments, where their potential sources are directly related to the use of the adjacent land (Tao *et al.*, 2021). An example of this are the lowland river catchments in agriculturally used areas, where the main source of pollution is a surface runoff from productive soils (Varekar *et al.*, 2021). In the case of larger river catchments, a number of potential pollution sources usually increases and consequently leads to difficulties in their identification and unambiguous interpretation (Zhang *et al.*, 2021).

As already mentioned, the identification of pollution sources is a multi-step process and requires a number of research studies dedicated to complex chemical analyses of water parameters. As a rule, an expansion of the number of parameters describing surface water quality contributes to an increase in the degree of accuracy of the pollution source identification process (Yuan and Liang, 2021). This regularity results from the fact that while extending the scope of monitoring studies, the amount of information on water quality increases (Zeunert and Meon, 2020), which can be used to describe phenomena and processes occur-

Fig. 1. The established network of monitoring points.

ring naturally in the environment or will allow to indicate parameters occurring in abnormal amounts at selected monitoring points (Wu *et al.*, 2020a). However, it should be emphasized that this approach leads to an increase in the complexity of the database and discredits the use of only simple methods for analysing the dataset even in the preliminary identification of pollution sources (Ma *et al.*, 2020). More complex environmental databases require the use of multivariate statistical inference methods that allow for the identification of the often non-linear relationships that exist between individual quantities measured in environmental matrices (Kaur *et al.*, 2020). Such methods of statistical analysis include methods of clustering variables using dendrograms or reduction methods, as a result of which it is possible to indicate factors that are somehow correlated with individual pollutants. Both the analysis of associations between variables depicted by dendrograms and the determination of factors allow relatively accurate identification of sources of migration of pollutants to surface waters (Yuan *et al.*, 2020).

Taking into account the relationships and transformations taking place in surface waters, the aim of the study was to identify sources of pollution in the waters of the Narew River catchment on the base of monitoring data collected by the General Inspectorate for Environmental Protection. In order to verify the research objective, the state of water in the Narew River catchment was assessed, similarities and differences between the various tributaries of the Narew River were identified, factors and processes responsible for transformations in the aquatic environment were identified, and finally the main sources of pollution in the catchment were identified.

RESEARCH METHODOLOGY

Area of research

The Narew River is among the longest rivers in Poland. Its total length is 484 km, of which 448 km are located in the Polish territory. The Narew River belongs to the lowland rivers (Skorbiłowicz, 2006). It forms numerous extensive valleys, bogs and marshes. Due to the fact that the Narew River consists of numerous branches, it is counted as anastomosing river (Skorbiłowicz, 2003). The entire catchment area covers an area of 75,175.2 km^2, of which 53,787 km^2 are located in Poland (Skorbiłowicz and Skorbiłowicz, 2009). The land use characteristics of the Narew River catchment is relatively complex. According to literature data, the dominant land uses are: arable land (38.56%), forests (41.87%), permanent grassland (12.36%) and wetlands (4.15%). Other land uses in the catchment area are rural buildings (1.16%), urban residential buildings (1.07%), artificial water bodies (0.58%), industrial and commercial areas (0.21%) and transport areas (0.04%) (Nasiłowska, 2008). On the other hand, the main sources of pollution in the Narew River catchment are due to the development and functioning of larger settlement units (Białystok, Łomża, Nowogród and Ostrołęka) located within the catchment. However, a quality of the aquatic environment of the Narew River and its tributaries may be affected by both anthropogenic factors and processes naturally occurring in surface water. Assessment of water quality and identification of pollution sources in the Narew River catchment was carried out using data obtained from 26 measurement points (Table 1), while Figure 1 presents the monitoring network.

Monitored parameters

The study uses results of research conducted within the framework of diagnostic monitoring conducted by the General Inspectorate for Environmental Protection in the area of the Narew River catchment in 2017 and 2018. For the purposes of pollution identification, a database was created in which a number of parameters describing water quality was taken into account. A total of 19 surface water quality parameters were included in the database. The individual parameters were analysed in accordance with the research methods and procedures developed by the General Inspectorate for Environmental Protection and the applicable standards for the measurement methodology of the environmental quantities analysed.

Among the physical parameters, colour, total suspended solids (TSS), total dissolved solids (TDS) and conductivity were included in the database. Chemical parameters included were carbon compounds expressed as biological oxygen demand (BOD), chemical oxygen demand (COD) and total organic carbon (TOC). Additionally, the database includes a number of parameters describing the amounts of selected nutrients, including total nitrogen (Total N), Total Kjeldahl Nitrogen (TKN), ammonia nitrogen, nitrate nitrogen (III) and nitrate nitrogen (V), phosphates (V), total phosphorus (Total P) and parameters describing water salinity in the form of chlorides and sulphates. Additionally, concentrations of selected macroelements were also included in the database, taking into account the amounts of calcium (Ca) and magnesium (Mg).

Table 1. Location of environmental sampling points.

River	Point of collection defined by the GIEP	Watercourse code
Szkwa	Rozóg	PLRW2000172651852
Pisa	Rygarby	PLRW700020584789
Biebrza	Stary Rogożyn	PLRW200023262151
Czarna	River mouth to the Narew	PLRW2000172613749
Rudnia	River mouth to the Narew	PLRW200017261369
Małynka	River mouth to the Narew	PLRW2000172613529
Ruda	River mouth to the Narew	PLRW200017261349
Olszanka	River mouth to the Narew	PLRW200023261312
Omulew	Grabowo	PLRW200019265499
Różanica	Prostki	PLRW2000172628969
Klusówka	Klusek	PLRW20001726719699
Narewka	Białowieża	PLRW200024261253
Łoknica	River mouth to the Narew	PLRW200017261389
Krzywa Noga	Jankowo Młodzianowo	PLRW200017265129
Czeczotka	Wojciechowice	PLRW200017265369
Bug	Wyszków	PLRW200021266979
Rządza	Krzywica	PLRW200017267167
Narew	Ostrołęka	PLRW20002126539
Narew	Ogony	PLRW20002126555
Narew	Pułtusk	PLRW20002126599
Narew	Suraż	PLRW200019261539
Narew	Ploski	PLRW200019261399
Narew	Siemianówka	PLRW200002611399
Narew	Babia Góra	PLRW200024261119
Narew	Nowogród	PLRW20002126399

STATISTICAL ANALYSIS

The analysis of factors that may affect water quality in the Narew River catchment was divided into 3 stages. In the first stage basic statistical values describing distribution of variables in the data set were analysed taking into account the arithmetic mean, standard deviation, minimum and maximum values and variability of particular variables. The analysis of these measures first allows the identification of parameters that deviate from the natural character of changes, resulting from seasonal changes in the environment (Skorbiłowicz *et al.*, 2017).

In the second stage of identification, taking into account the number of variables included in the database and the number and complexity of phenomena that can be described with their use, a principal component analysis with factor rotation using the standardized Varimax method was used. Using this type of analysis, factors responsible for shaping surface water quality in the Narew River catchment were identified. The selection of the number of factors was based on the Kaiser criterion, according to which the eigenvalue of a single factor should be greater than 1. This means that one extracted factor explains more changes than a single variable included in the analysis. According to the described methodology, no more than 5 factors should be used to interpret changes. This number of distinguished factors is also in line with the Cattell criterion (Fig. 2), which allows to accept for verification the factors that explain more than 5% of the changes occurring in the data set. However, interpretation of such a large number of factors in the adopted set of variables did not allow unambiguous identification of the pollution sources. Therefore, only 2 factors determining the largest percentage of changes in the data set were included in further descriptions. For the purposes of factor identification, it was assumed that the variables included in its description would be characterised by a factor load modulus greater than or equal to the value of 0.70 (Skorbiłowicz, 2013, 2016).

The next phase of statistical analysis was the use of clustering by the Ward's method, which was used to identify watercourses in the Narew River catchment area where similar changes in the chemical composition of water occur. All variables adopted for the analysis were subjected to a standardization procedure in order to obtain normalized values of individual variables. Taking into account the fact that the values included in the statistical analyses are expressed in different units, this approach allowed to increase the precision of the analyses. The described methods have been gaining increasing interest in recent years due to their universal character. Additionally, they have been successfully applied to interpret changes in surface watercourses from different regions of the world (Bakure *et al.*, 2020; Ma *et al.*, 2020; Cui *et al.*, 2021; Tao *et al.*, 2021).

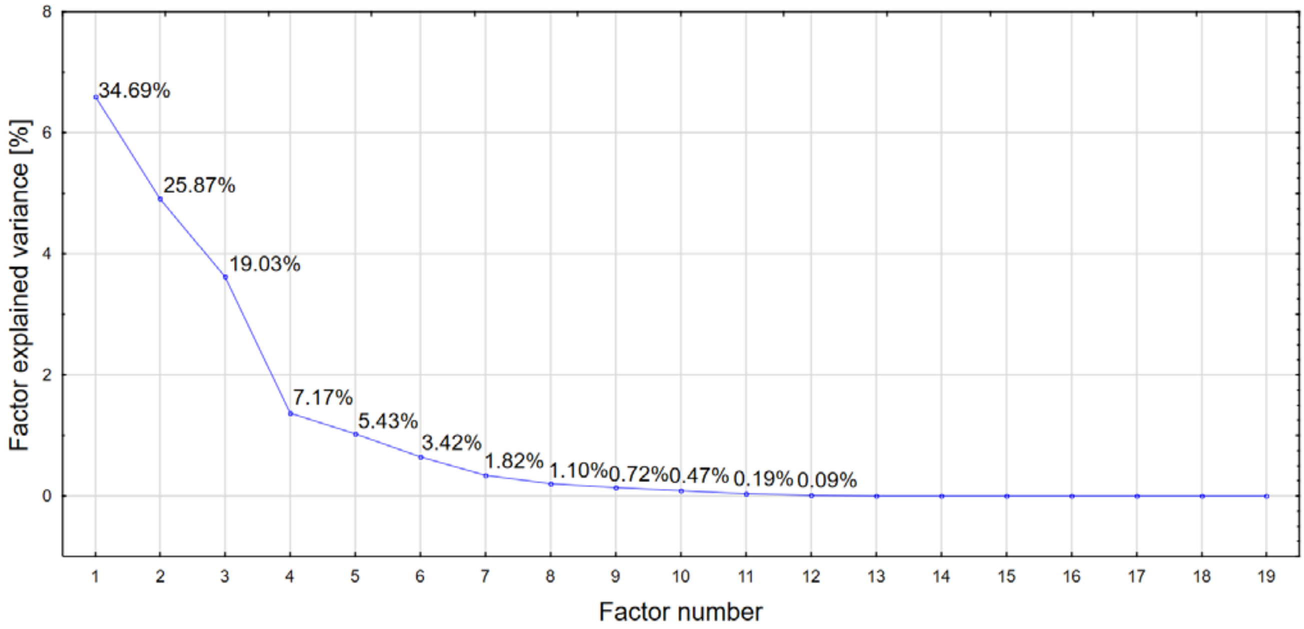

Fig. 2. Scatter plot and Kaiser criterion for factor analysis.

RESEARCH RESULTS AND DISCUSSION

Among the considered variables, the values of pH, conductivity, TDS and Ca were characterized by low variability. The range of variability for the remaining parameters was in the range of average, strong and very strong variability. This observation may indicate that in the Narew River catchment area, surface water quality is more dependent on external factors that may be responsible for the inflow of pollutants and at the same time disturb the natural transformations occurring in surface waters as a result of seasonality especially this is evident in nutrient compounds (Lee *et al.*, 2018; Yan *et al.*, 2018; Chen *et al.*, 2020). Similar observations are indicated by measures of the spread of the data set and values of arithmetic means and standard deviations, where the largest differences between minimum and maximum concentrations were observed for the studied forms of nitrogen and phosphorus.

The quality of surface water in the Narew River catchment may be affected by the relatively large area, the catchment cover and the relatively varied land use. Additionally, the presence of pollution sources is indicated by the results of statistical analysis, where coefficients of variation and values of arithmetic means, standard deviations, minimum and maximum values observed in the whole catchment were compared (Table 2).

Identification of factors responsible for shaping surface water quality in the Narew River catchment was based on the principal component analysis (Table 3). As a result of the analysis. two factors were identified. The first of these consisted mainly of conductivity values, TDS, SO_4^{2-}, Ca, Mg, N-NO_3, N-NO_2 and total N. However, it should be stressed that potential sources of nitrogen forms in the aquatic environment may also be the surface runoff or the dominant catchment land use (Haymale *et al.*, 2020). From this fact, it may follow that in the discussed cluster of variables associations with particular aerobic forms of nitrogen and with total nitrogen were observed (Dzierzbicka-Głowacka *et al.*, 2019; Krevs *et al.*, 2019; Szatten and Habel, 2020). According to the literature (Farid *et al.*, 2020; Marques *et al.*, 2020; Yin *et al.*, 2020), nitrogen quantities in river catchments are determined by the amount of surface runoff from agricultural land, especially during plant vegetation periods. Additionally, surface runoff can result in the de-

Table 2. Descriptive statistics of the parameters studied in the waters of the Narew River catchment.

Variable	Average mean	Standard deviation	Min	Max	Variation
pH	–	–	7.25	8.40	3.20
Color	54.42	32.41	19.00	146.50	54.01
TSS	10.73	10.52	3.00	44.20	98.27
BOD	2.61	1.16	1.20	6.77	44.64
TOC	12.52	5.50	5.20	26.80	43.62
COD_{Cr}	37.25	13.88	16.77	68.00	40.52
Conductivity	420.58	85.41	234.67	588.00	18.92
TDS	308.32	60.59	219.00	460.00	19.01
SO_4^{2-}	28.54	10.10	14.70	45.43	35.60
Cl^-	12.78	6.53	2.75	26.92	52.83
Ca	75.00	10.75	58.50	103.00	16.17
Mg	10.91	3.44	5.34	17.70	32.97
N-NH_4	0.14	0.18	0.03	1.00	136.20
TKN	1.35	0.40	0.85	2.54	32.31
N-NO_3	1.19	1.45	0.07	8.00	117.03
N-NO_2	0.01	0.01	0.00	0.05	69.98
Total N	2.54	1.44	1.26	9.00	60.39
P-PO_4	0.07	0.10	0.02	0.58	37.52
Total P	0.14	0.06	0.08	0.43	36.39

Table 3. Results of the factor analysis.

Variable	Factor 1	Factor 2
Color	-0.54	0.14
TSS	0.15	0.86
BOD	-0.06	0.88
TOC	-0.53	0.39
COD_{Cr}	-0.57	0.58
Conductivity	0.92	0.17
TDS	0.87	0.09
SO_4^{2-}	0.80	0.36
Cl^-	0.51	0.63
Ca	0.70	0.04
Mg	0.72	-0.51
pH	-0.09	0.42
$N\text{-}NH_4$	0.19	0.19
TKN	-0.20	0.75
$N\text{-}NO_3$	0.83	-0.13
$N\text{-}NO_2$	0.82	-0.09
Total N	0.80	0.05
$P\text{-}PO_4$	0.06	0.60
Total P	0.04	0.96
Explained variance	34.69	25.87

livery of dissolved substances, suspended solids and Ca and Mg compounds (Bakure *et al.*, 2020; Siegwald and de Jong, 2020; Wu *et al.*, 2020). In addition, the TDS value directly describes the amount of solutes leached from the soil substrate that may be carried with the rainwater flow (Leroy *et al.*, 2016; Araujo and Neto, 2018; Cockerill *et al.*, 2019; Trajkovic *et al.*, 2020; Wang *et al.*, 2020). Based on the described relationships, factor 1 can be associated with surface runoff and explains 34.69% of the changes in water quality in the Narew River catchment.

The second factor included TSS, BOD, TKN and total P. The variables included in this cluster can be related to the supply of treated wastewater (Campo *et al.*, 2020; Nhut *et al.*, 2020), in which usually carbon compounds described by the parameter COD, BOD or TOC and nitrogen and phosphorus compounds are present (We *et al.*, 2020). A particularly important variable in this cluster is BOD and total P. which indicate the inflow of carbon and phosphorus compounds (Kang *et al.*, 2020). The specific nature of the biological wastewater treatment plant operation does not guarantee complete degradation of carbon compounds (Letshwenyo *et al.*, 2020). Consequently, this leads to migration of the pollutants present in wastewater to the aquatic environment (Arias *et al.*, 2020; Boujelben *et al.*, 2020). In addition to carbon compounds, the treated wastewater may also contain residual organic forms of nitrogen and ammonium nitrogen collectively described as TKN (Petrovski *et al.*, 2020). Which, as a result of continuous delivery to the aquatic environment, may clearly indicate the impact of this type of facility on the quality of receiver water (Nguyen *et al.*, 2020). On the other hand, the lack of association with other forms of nitrogen can be justified by the course of biological wastewater treatment itself, during which individual forms of nitrogen are removed with relatively high efficiency. Hence, the concentrations of $N\text{-}NO_3$, $N\text{-}NO_2$ and total N are lower compared to the concentrations of these compounds in surface water (Mohtadi *et al.*, 2020; Rahman *et al.*, 2020). Based on the given interpretation, the factor 2 can be identified as the influence of effluent from wastewater treatment plants on the quality of receiver water, which are ultimately watercourses in the Narew River catchment. This factor explained 25.87% of the changes observed in the entire catchment.

Verification of the identified factors was based on the value of factor loadings at individual sampling points (Fig. 3). On this basis, it is possible to indicate points where the greatest intensification of influence of a given factor on changes in the Narew River water is noted. The factor 1, which was defined as the surface runoff, had the highest

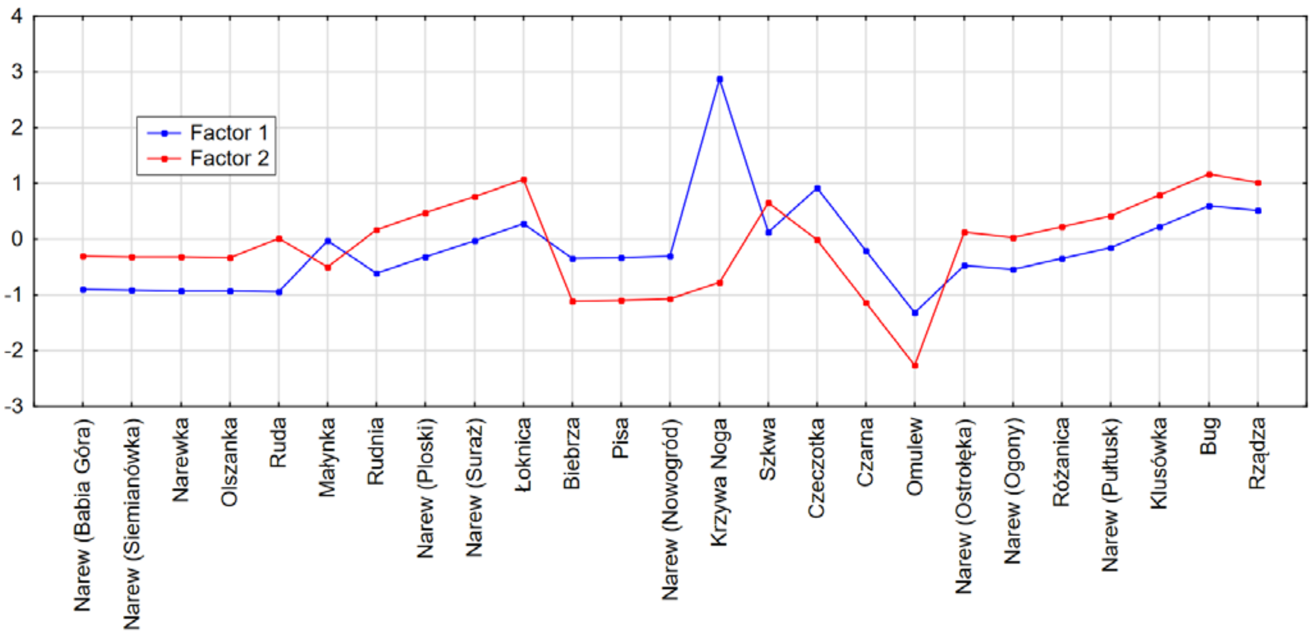

Fig. 3. Distribution of factor loads in the Narew River catchment area.

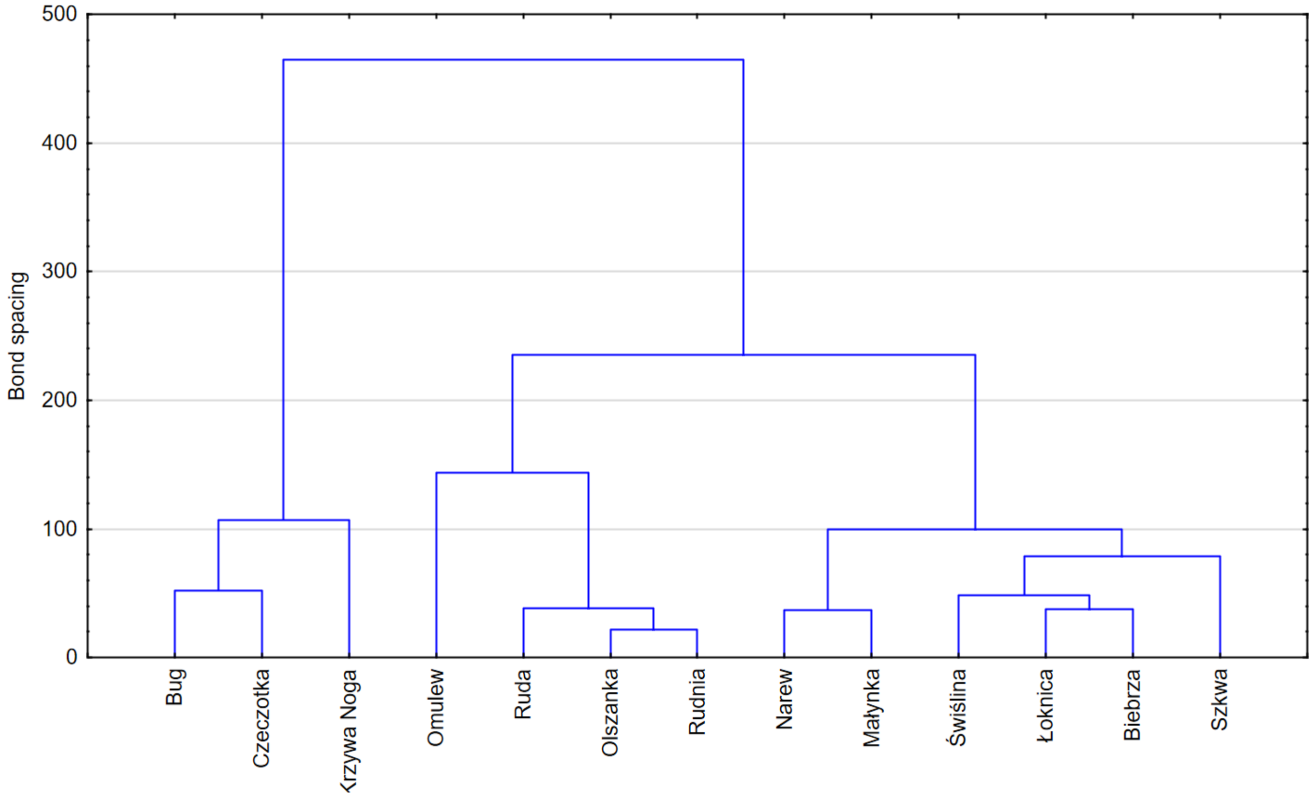

Fig. 4. Cluster analysis of rivers in the Narew River catchment area.

value of factor load at the point Krzywa Noga, while up to this point the value of the load gradually increased. This indicates an increasing contribution of surface runoff to water quality in the catchment from Babia Góra to Krzywa Noga. In the case of the factor 2 responsible for the inflow of treated sewage to the waters of the catchment, four points were observed where it took extreme values. At the Łoknica point, this value can be associated with wastewater inflow from rural households, while the Szkwa is a relatively large river receiving treated wastewater from several potential sources. On the other hand, the factor load values at Ostrołęka and Bug can be directly correlated with the functioning of WWTPs present in the agglomeration area where Narew and Bug Rivers flow.

Figure 4 shows results of a cluster analysis conducted using the Ward's method where individual tributaries of the Narew River were grouped. The aim of this approach was to identify watercourses with similar chemical characteristics. According to the results of the analysis, 2 clusters of flowing surface waters with similar quality characteristics were identified. In the first cluster the rivers Bug, Czeczotka and Krzywa Noga were included. Interpretation of the reason of connection of these rivers in a single cluster may result from values of the factor 2 loadings (Fig. 3), which took extreme values within measurement points located on these rivers. In contrast, the other studied tributaries of the Narew River were placed in a separate cluster. This may be due to the fact that the dominant factor shaping surface water quality in the Narew River catchment is the surface runoff. Hence, this cluster corresponds to rivers, chemical characteristics of which are shaped by the surface runoff.

CONCLUSIONS

The largest range of variation in the data set was observed for nutrients and suspended solids. This may be due to the fact that the Narew River catchment is predominantly used for agricultural purposes and, in addition, it has a relatively large number of potential anthropogenic sources. Analysis of the basic statistical measures describing the data set showed that the studied quality parameters in the waters of the Narew River catchment were mostly characterized by strong and very strong variability. This observation may indicate that the transformations of the individual parameters studied are not the result of natural phenomena occurring in surface waters, but are dependent on external factors.

Despite the high level of complexity of the database, the methods of advanced statistical inference applied made it possible to determine the factors affecting surface water quality in the Narew River catchment area. The analyses showed that water quality in the Narew River catchment is affected by surface runoff and inflow of treated wastewater.

Based on the values of the factor loads at particular points of the monitoring network, sampling sites were indicated where the identified factors showed the greatest intensification of impact. In the case of surface runoff, the greatest impact was observed at the Krzywa Noga and Czeczotka points, while in the case of treated wastewater inflow the greatest impact was observed at Łoknica, Szkwa and Bug. These observations coincided with the results of agglomeration carried out for individual rivers.

The identification of pollution sources in the Narew River catchment area indicates that in order to improve water quality, the fertilizer management of agricultural areas in the catchment area should be regulated and the efficiency of the wastewater treatment systems should be improved.

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