QUALITATIVE ANALYSIS AND STATISTICAL MODELS BETWEEN SPRING WATER QUALITY INDICATORS IN ALBA COUNTY, ROMANIA

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Abstract

Based on contemporary environmental issues, related to water scarcity, the research outlines the current situation regarding the quality of spring waters in Alba County. Spring water is an alternative that is less and less investigated and exploited if it is a free, natural, untreated source. The quality of spring drinking water is regulated by Law no. 458/2002 (transposing the Directive 98/83/EC on the quality of drinking water). Five areas with 132 water sources were monitored, and for the most representative and polluted springs, their monitoring (physico-chemical and microbiological indicators) was performed for a period of 3 years. Most sources are microbial contaminated regardless of the season or the water catchment and spring arrangement. There is also a diffuse microbiological pollution in some localities, which indicates that the phenomena of natural purification no longer occur. It was found that in Alba County the percentage of drinking spring water sources is relatively low as follows: Alba Iulia-Teiuş area - 37%, Sebeş-Cugir - 48%, Câmpeni-Zlatna - 72%, Blaj - 25%, Aiud-Ocna Mureş - 20%. The correlative analysis shows that between the total viable count and other physico-chemical parameters (ammonium, nitrates and nitritesions) of spring water there are appreciable correlations. The correlation coefficients between these parameters vary between 0.67 and 0.92. The equations of the statistical models can approximate the variation in time of the microbial growth. These represent a control and prediction tool for the appreciation of the spring water quality in time, knowing only the physico-chemical parameters.

Key words: contamination, nitrogen cycle, quality, spring water, statistics.

INTRODUCTION

According to the Drinking Water Directive (Directive 2000/60), water is not a commercial good, but a heritage that must be protected, and treated as such. Water is a fundamental social requirement and is an essential "element" for the maintenance and development of life on our planet (Dinka, 2018).

Water, although it was thought to be an inexhaustible gift of nature, nowadays there is a water crisis, especially at the regional level. It is estimated that around 2020-2025 the water crisis will manifest itself globally (Jelev, 2008). Clean drinking water is an essential element in ensuring public health and quality of life. Water registers various sources of contamination (Ecoaqua, 2002).

Public water systems use only surface water (lakes, rivers). Spring water is important

because these sources can be bottled and traded (Directive 2009/54). The consumption of spring and well water appears as a new situation generated by the idea that it is healthier than tap water and is free (Glevitzky & Popa, 2012).

The potability of the water is given by its quality indicators. The Romanian Law no. 311/2004 (L.311, 2004) which modify and revise the Law no. 458/2002 (L.458, 2002) on drinking water quality, it isharmonized with the EU legislation (Directive 98/83/EC) on the quality of water intended for human consumption.

The paper tests the quality of spring waters in Alba County. The results of the study show the variation of the physico-chemical and microbiological parameters for these waters. At the same time, the interdependence of these parameters over time is determined using mathematical models.

The purpose of this paper is to investigate the quality of five spring waters in Alba County, Romania. The results of the study show that the nitrate content and microbiological parameters have values that exceed the limits for drinking water.

MATERIALS AND METHODS

In a first stage, an analysis of the data related to Alba County for public groundwater sources was used. Thus, in 2017, several 132 public water sources were monitored. For a better data management, the county was divided into 5 zones, as follows: Area I: Alba Iulia - Teius: Area II: Sebeș - Cugir; Area III: Cîmpeni -Zlatna; Area IV: Blaj and Area V: Aiud - Ocna Mures. For the relevance of the results, physicochemical and microbiological parameters were investigated according to European regulations. Three physico-chemical parameters of the water were monitored in the period 2017-2019 on the most polluted spring in each area. These are: ammonium, nitrates and nitrites content. The methods of the rapid spectrophotometric determinations involve the use of spectrophotometer Spectroquant NOVA 60 and SQ specific kits.

The determined microbiological parameters are: total number of bacteria growing 37°C (SR EN ISO 6222, 2004), detection and counting of *Escherichia coli* and coliform bacteria Part 1: Membrane filtration method (SR EN ISO 9308-1, 2004; SR EN ISO 7889-2, 2002) and identification and counting of intestinal enterococci Part 2: Membrane filtration method (SR ISO 21528-1/2, 2004). The statistical analysis of the obtained data can provide information related to the evolution in time of the most important water parameters.

RESULTS AND DISCUSSIONS

During 2017-2019, a number of 132 public water sources from Alba County were monitored. Of these, only 38 sources were potable. The other 94 have one or more physico-chemical and microbiological parameters above the limits.

In Figure 1 are depicted the results of the analyses related to quality indicators of water sources from Alba County for 2017.

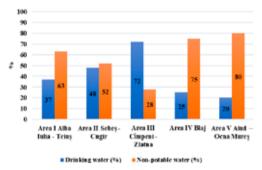


Figure 1. Quality of public groundwater sources in Alba County for the 5 areas

The analysis of the obtained results (Figure 1) shows that most investigated of the groundwater sources (springs) are not drinkable. The highest percentage of nonpotability is found in area V - Aiud-Ocna Mures (80%), and the lowest in area III -Câmpeni-Zlatna (28%). At the same time, it is observed that in the mountain region (area III) has provided safe, high-quality water compared to the other regions. The mountainous region (Câmpeni area) is the closest to the drinking water standard in Alba County.

Living organisms, dead organic matter, mineral and organic compounds dissolved in an aquatic ecosystem are never in an inert state. They are in a permanent transformation and circulation that conditions the existence of living systems. Thus, complex relationships are created between the components of the ecosystem that ensure the evolving stability of these components or the balance of the entire structure.

Nitrogen, an important nutrient in aquatic ecosystems, enters in water in several ways and is found in many forms: molecular nitrogen, nitrogen oxides, ammonia, ammonium, nitrites and nitrates.

In the ecosystem, nitrogen enters the biogeochemical circuit, determined by an interactional complex of factors in the aquatic ecosystem. Algae can use both free nitrogen from water and ammoniacal salts (NH₃) and, after depletion, nitrate-nitrogen (NO₃⁻) (Botnariuc and Vădineanu, 1982). Bacteria plays vital role in the circulation of nitrogen in the specific circuit of aquatic ecosystems. The phases of bacterial transformations of nitrogen compounds are reversible, the direction of the

processes being dependent mainly on the concentration of dissolved oxygen.

Water samples were analysed in time, monthly, from January 2017 to December 2019. Experimental data obtained from the analysis of physico-chemical parameters over 3 years are presented comparatively for the representative sources in the five areas in Figures 2-4.

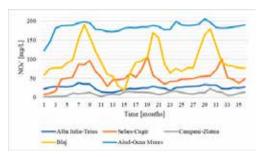


Figure 2. Variation of nitrate content in time for the 5 springs

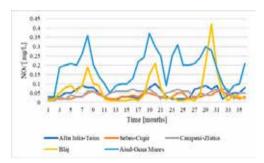


Figure 3. Variation of nitrite content in time for the 5 springs

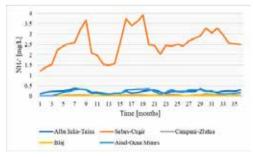


Figure 4. Variation of ammonium ion content in time for the 5 springs

Nitrites are present in water due to pollution with organic substances, either by partial oxidation of the amino radical or by reduction of nitrates. Their presence indicates an older pollution, but together with high concentrations of ammonium ions show that the pollution is continuous (Glevitzky et al., 2020).

Ammonium ions appear because of water pollution with various organic substances, which decompose, being the first stage of degradation of nitrogenous substances. Its presence indicates recent pollution.

The experimental data obtained from the analysis of microbiological parameters over a period of three years are presented comparatively for the representative sources in the five areas in Figures 5-8.

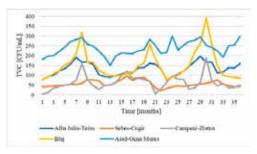


Figure 5. Variation of the total number of mesophilic aerobic bacteria in time for the 5 springs

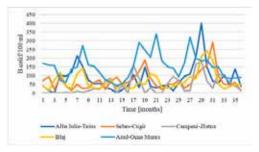


Figure 6. Variation of the number of coliform bacteria in time for the 5 springs

It is found that water from springs located in different areas has a high microbial load. The plain areas of Aiud-Ocna Mureş, Alba Iulia-Teiuş, Blaj and Sebeş have a higher microbiological load, with large variations depending on the season. For mountain and hill areas, there are small fluctuations in microbiological parameters over time.

An increased number of mesophilic aerobic bacteria indicate a risk of waterborne pathogens (bacteria, viruses, fungi, parasites). As a result of exceeding the allowed limits for these parameters, it can be considered that the

springs are polluted and due to infiltrations from wastewater, manure, etc. (Glevitzky et al., 2020).

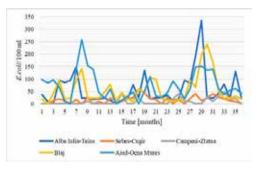


Figure 7. Incidence of E. coli bacteria in time for the 5 sources

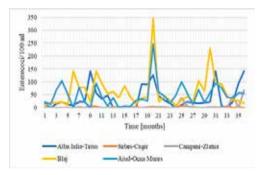


Figure 8. Incidence of intestinal enterococci in time for the 5 sources

Escherichia coli can be transmitted through water consumption and causes gastroenteritis and sepsis in children and young people of different animal species (Mănescu, 1989). E. coli can survive in drinking water from wells for 6 months and in canal water for 4 months (Drăghici, 2002). There is also a diffuse microbiological pollution in some localities, which indicates that the groundwater is polluted and therefore the phenomena of natural purification undergo transformations; nature no longer manages, by its own means, to fulfil its role of purifier.

In many cases the connection between two or more parameters that describe a particular process is close enough that the variation of one parameter can be controlled and expressed based on the variation of the other parameters. Functional links of this kind are called stochastic or probabilistic links. The study of such links has led to the development of multiple correlation theory (Savii and Luchin, 2000).

To develop statistical models, the influence of two parameters was assessed by performing a multiple correlation analysis. The microbial load (aerobic bacteria) was the dependent variable, respectively the content of nitrates. nitrites and ammonium ions over time, being independent variables. Starting from the nitrogen cycle in water - the dependence of the total number of germs depending on the presence of ammonium ions, nitrates or nitrites over time, as case studies for these statistical mathematical models were analysed for 3 years per month the most polluted sources of spring water from the 5 areas of Alba County.

To describe as accurately as possible, the dependence between nitrates, nitrites, ammonium content, and microbial load over time, it was desired to obtain a second order polynomial equation. Equation (1) represents the general form of mathematical models that describe the second order polynomial correlation.

$$y=a_0+a_1x_1+a_2x_2+a_3x_1x_2+a_4x_1^2+a_5x_1^2$$
 (1)

In equation (1) ai represents the coefficients of the equation, and the variables x_1 , x_2 and ycorrespond to the following parameters:

y - total number of aerobic bacteria, log [cfu/mL];

 $x_1 - NO_3$, NO_2 , NH_4 content [mg/L];

x₂ - time [months].

Using the Matlab® program, the system of equations was solved and, the experimental data were processed and analysed. equations of the statistical models obtained based on the multiple regression are presented in Table 1. They are valid on the studied value range.

Table 1. Equations of the statistical models obtained in the case of the spring from Area I - Alba Iulia-Teius

| Variable | Equations of statistical models |
|--|---|
| | l |
| Nitrates (NO ₃ -) | $y=1.891-0.002 \cdot x_{1}+0.004 \cdot x_{2}+1.777 \cdot 10^{-4} \cdot x_{1} \cdot x_{2}+2.662 \cdot 10^{-4} \cdot x_{1}^{2}-1.598 \cdot 10^{-4} \cdot x_{2}^{2}$ |
| Nitrites (NO ₂ -) | $y=1.796 + 7.060 \cdot x_1 + 0.006 \cdot x_2 - 0.055 \cdot x_1 \cdot x_2 - 23.232 \cdot x_1^2 - 6.020 \cdot 10^{-5} \cdot x_2^2$ |
| Ammonium (NH ₄ ⁺) | $y=1.750+1.279 \cdot x_1+0.006 \cdot x_2+0.007 \cdot x_1 \cdot x_2-0.06 \cdot x_1^2-1.522 \cdot 10^{-4} \cdot x_2^2$ |

The experimental data and the surfaces generated by the statistical models for Area I - Alba Iulia-Teiuş are presented in Figures 9 to 11.

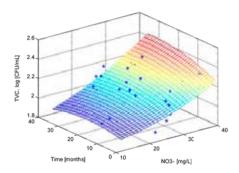


Figure 9. Variation of the microbial load depending on the nitrate content and time for the spring in Area I -Alba Iulia-Teius

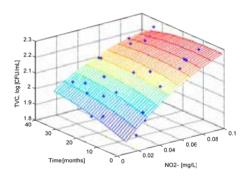


Figure 10. Variation of the microbial load depending on the nitrite content and time for the spring in Area I - Alba Iulia-Teius

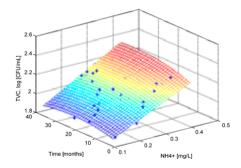


Figure 11. Variation of the microbial load depending to the ammonium ion content and time for the spring in Area I - Alba Iulia-Teiuş

After calculating the model coefficients, it is necessary to make a comparison between the model predictions and the data provided by the process. Dispersion σ^2 , standard deviation σ , correlation coefficient R, accuracy coefficient R^2 were used as indicators of model adequacy (Table 2).

Table 2. Adequacy indicators of the determined statistical models

| Variable | σ^2 | σ | \mathbb{R}^2 | R |
|----------|------------|-------|----------------|-------|
| Nitrates | 0.004 | 0.065 | 0.654 | 0.808 |
| Nitrites | 0.002 | 0.043 | 0.847 | 0.920 |
| Ammonium | 0.003 | 0.060 | 0.716 | 0.846 |

The values of the concordance indicators argue for a good capacity to predict statistical models. The experimental data together with the surfaces generated by the statistical mathematical models for Area II - Sebeş-Cugir are presented in Figures 12-14.

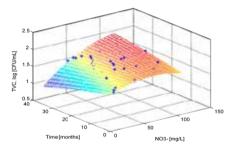


Figure 12. Variation of the microbial load depending on the nitrate content and time for the spring in Area II -Sebeş-Cugir

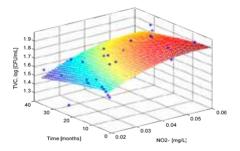


Figure 13. Variation of the microbial load depending on the nitrite content and time for the spring in Area II -Sebeş-Cugir

The equations of the statistical models obtained after the nonlinear multiple regression are presented in Table 3. They are valid on the studied value range.

Table 3. Equations of the statistical models obtained in the case of the spring from Area II - Sebeş-Cugir

| Variable | Equations of statistical models |
|--|---|
| Nitrates (NO ₃ -) | $y = 1.520 + 0.009 \cdot x_1 - 0.015 \cdot x_2 + 2.498 \cdot 10^{-4} \cdot x_1 \cdot x_2 - 7.052 \cdot 10^{-5} \cdot x_1^2 - 6.262 \cdot 10^{-5} \cdot x_2^2$ |
| Nitrites (NO ₂ -) | $y = 1.314 + 21.790 \cdot x_1 - 0.003 \cdot x_2 + 0.015 \cdot x_1 \cdot x_2 - 198.024 \cdot x_1^2 - 4.505 \cdot 10^{-5} \cdot x_2^2$ |
| Ammonium (NH ₄ ⁺) | $y = 1.730 - 0.057 \cdot x_1 - 0.010 \cdot x_2 + 0.006 \cdot x_1 \cdot x_2 + 0.020 \cdot x_1^2 - 2.749 \cdot 10^{-4} \cdot x_2^2$ |

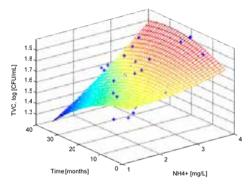


Figure 14. Variation of the microbial load depending to the ammonium ion content and time for the spring in Area II - Sebeş-Cugir

In order to compare the model prediction and the data provided by the actual process, the suitability indicators of the model presented in Table 4 were calculated.

Table 4. Adequacy indicators of the determined statistical models

| Variable | σ^2 | σ | \mathbb{R}^2 | R |
|----------|------------|-------|----------------|-------|
| Nitrates | 0.008 | 0.091 | 0.564 | 0.751 |
| Nitrites | 0.011 | 0.103 | 0.449 | 0.670 |
| Ammonium | 0.010 | 0.097 | 0.505 | 0.711 |

Results of the concordance indicators for the statistical models confirm appreciable correlations between the studied parameters. The experimental data together with the surfaces generated by the statistical mathematical models for Zone III - Câmpeni - Zlatna are presented in Figures 15-17. The equations of the statistical models obtained

following the nonlinear multiple regression of

order 2 are presented in Table 5. They are valid on the studied value range.

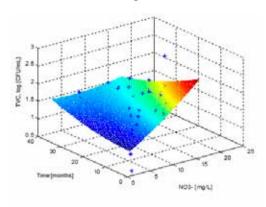


Figure 15. Variation of the microbial load depending on the nitrate content and time for the spring in Area III - Câmpeni-Zlatna

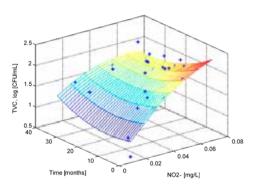


Figure 16. Variation of the microbial load depending on the nitrite content and time for the spring in Area III – Câmpeni-Zlatna

Table 5. Equations of the statistical models obtained in the case of the spring from Area III - Câmpeni-Zlatna

| Variable | Equations of statistical models |
|--|---|
| Nitrates (NO ₃ -) | $y = 1.089 + 0.078 \cdot x_1 + 0.0036 \cdot x_2 - 0.0027 \cdot x_1 \cdot x_2 + 0.0015 \cdot x_1^2 + 3.3753 \cdot 10^{-4} \cdot x_2^2$ |
| Nitrites (NO ₂ -) | $y = 0.592 + 42.040 \cdot x_1 - 0.011 \cdot x_2 - 0.424 \cdot x_1 \cdot x_2 - 219.871 \cdot x_1^2 + 6.406 \cdot 10^{-4} \cdot x_2^2$ |
| Ammonium (NH ₄ ⁺) | $y = 0.685 + 26.003 \cdot x_1 + 0.040 \cdot x_2 - 0.856 \cdot x_1 \cdot x_2 + 44.453 \cdot x_1^2 - 3.244 \cdot 10^{-4} \cdot x_2^2$ |

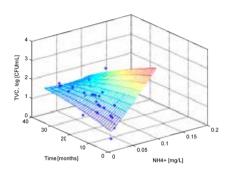


Figure 17. Variation of the microbial load depending to the ammonium ion content and time for the spring in Area III – Câmpeni-Zlatna

The adequacy indicators of the statistical models obtained by comparing the model predictions with the data provided by the actual process are presented in Table 6.

Table 6. Adequacy indicators of the determined statistical models

| Variable | σ^2 | σ | \mathbb{R}^2 | R |
|----------|------------|-------|----------------|-------|
| Nitrates | 0.049 | 0.221 | 0.574 | 0.757 |
| Nitrites | 0.039 | 0.197 | 0.660 | 0.812 |
| Ammonium | 0.048 | 0.219 | 0.583 | 0.763 |

In the case of polynomial equations of the second order, the correlation coefficients prove a functionally satisfactory relationship between the investigated variables.

The experimental data together with the surfaces generated by the statistical mathematical models for Zone IV - Blaj are presented in Figures 18-20.

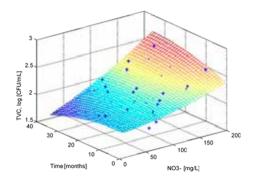


Figure 18. Variation of the microbial load depending on the nitrate content and time for the spring in Area IV - Blaj

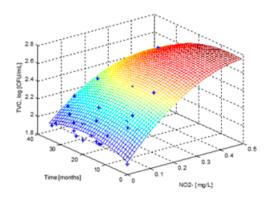


Figure 19. Variation of the microbial load depending on the nitrite content and time for the spring in Area IV - Blaj

The equations of the statistical models obtained from the nonlinear multiple regression are presented in Table 7 and the adequacy indicators in Table 8.

Table 7. Equations of the statistical models obtained in the case of the spring from Area IV - Blaj

| Variable | Equations of statistical models |
|--|---|
| Nitrates (NO ₃ -) | $y = 1.946 + 4.329 \cdot 10^{-4} \cdot x_1 + 8.790 \cdot 10^{-5} \cdot x_2 + 9.191 \cdot 10^{-5} \cdot x_1 \cdot x_2 + 7.895 \cdot 10^{-6} \cdot x_1^2 - 2.396 \cdot 10^{-4} \cdot x_2^2$ |
| Nitrites (NO ₂ -) | $y = 1.881 + 3.217 \cdot x_1 + 0.013 \cdot x_2 - 0.018 \cdot x_1 \cdot x_2 - 2.999 \cdot x_1^2 - 3.147 \cdot 10^{-4} \cdot x_2^2$ |
| Ammonium (NH ₄ ⁺) | $y = 2.193 - 28.933 \cdot x_1 + 0.007 \cdot x_2 + 0.051 \cdot x_1 \cdot x_2 + 1239.3 \cdot x_1^2 - 2.709 \cdot 10^{-4} \cdot x_2^2$ |

Table 8. Adequacy indicators of the determined statistical models

| Variable | σ^2 | σ | \mathbb{R}^2 | R |
|----------|------------|-------|----------------|-------|
| Nitrates | 0.005 | 0.071 | 0.843 | 0.918 |
| Nitrites | 0.007 | 0.082 | 0.793 | 0.890 |
| Ammonium | 0.012 | 0.109 | 0.635 | 0.797 |

The values of the correlation coefficients are appreciable, which indicates a good correlation in the case of multiple 2nd order regression.

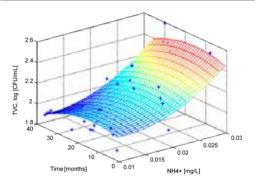


Figure 20. Variation of the microbial load depending to the ammonium ion content and time for the spring in Area IV - Blaj

The experimental data together with the surfaces generated by the statistical mathematical models for Zone V - Aiud-Ocna Mures are presented in Figures 21-23.

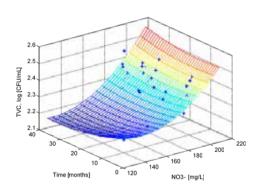


Figure 21. Variation of the microbial load depending on the nitrate content and time for the spring in Area V - Aiud-Ocna Mureş

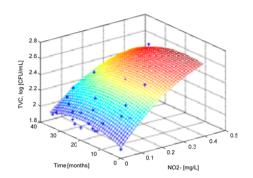


Figure 22. Variation of the microbial load depending on the nitrite content and time for the spring in Area V - Aiud-Ocna Mures

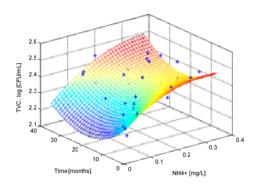


Figure 23. Variation of the microbial load depending to the ammonium ion content and time for the spring in Area V - Aiud-Ocna Mureş

The equations of the statistical models obtained from the nonlinear multiple regression are presented in Table 9.

Table 9. Equations of the statistical models obtained in the case of the spring from Area V - Aiud-Ocna Mureș

| Variable | Equations of statistical models |
|--|---|
| Nitrates (NO ₃ -) | $y = 1.946 + 4.329 \cdot 10^{-4} \cdot x_1 + 8.790 \cdot 10^{-5} \cdot x_2 + 9.191 \cdot 10^{-5} \cdot x_1 \cdot x_2 + 7.895 \cdot 10^{-6} \cdot x_1^2 - 2.396 \cdot 10^{-4} \cdot x_2^2$ |
| Nitrites (NO ₂ -) | $y = 1.881 + 3.217 \cdot x_1 + 0.013 \cdot x_2 - 0.018 \cdot x_1 \cdot x_2 - 2.999 \cdot x_1^2 - 3.147 \cdot 10^{-4} \cdot x_2^2$ |
| Ammonium (NH ₄ ⁺) | $y = 2.193 - 28.933 \cdot x_1 + 0.007 \cdot x_2 + 0.051 \cdot x_1 \cdot x_2 + 1239.3 \cdot x_1^2 - 2.709 \cdot 10^{-4} \cdot x_2^2$ |

Table 10. Adequacy indicators of the determined statistical models

| Variable | σ^2 | σ | \mathbb{R}^2 | R |
|----------|------------|-------|----------------|-------|
| Nitrates | 0.002 | 0.041 | 0.659 | 0.812 |
| Nitrites | 0.007 | 0.082 | 0.793 | 0.890 |
| Ammonium | 0.002 | 0.041 | 0.645 | 0.803 |

Calculated correlation parameters argue for a good prediction capacity of mathematical models. The performed analyses show that one or all the determined parameters, in most spring water samples, are well above the maximum limit allowed by law.

CONCLUSIONS

The research outlines the current situation regarding the quality of spring waters in Alba County. If public drinking water supply systems use only surface water (lakes, rivers),

spring water is a less and less investigated and exploited alternative even if it is a free, natural, untreated source of particular importance.

Five areas with 132 water sources from Alba County were monitored. The most representative and polluted sources were monitored (physico-chemical microbiological indicators) for a period of 3 years. It was observed that most of them were microbiologically polluted. It was found that in Alba County the percentage of drinking spring water sources is relatively low as follows: Alba Iulia-Teiuș (I) area - 37%, Sebeș-Cugir (II) -48%, Câmpeni-Zlatna (III) - 72%, Blaj (IV) -25%, Aiud-Ocna Mures (V) - 20%.

The equations of the statistical models obtained can be used as prediction models. With their help, the microbial load in the spring waters can be approximated, knowing the time of year when the sample is collected and the content of nitrates, nitrites, respectively the ammonium ion content of the sample. Correlation parameters argue for a good prediction capacity of statistical mathematical models.

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