APPLICATION OF COMPLEX STATISTICAL DISTRIBUTIONS AND NATURAL ISOTOPES OF HYDROGEN AND OXYGEN FOR ASSESSMENT OF WATER ORIGIN IN ŠAR MOUNTAINS AQUATORIUM

by

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This study considers the possibility of applying natural isotopes of water, tritium, and oxygen ^{18}O, to analyze the connection between groundwater, surface, and precipitation waters. This analysis also enables the determination of the age of groundwater, separated from the cycle of water circulation in nature. Based on these methods, it is possible to reliably determine, by chronologically accurate method, the possibility of industrial application and application for human needs of the waters of this aquatorium.

Key words: Šar mountains aquatorium, tritium, oxygen ^{18}O

INTRODUCTION

More recently, the efforts have been made to develop control, protection and enhancement of water resources, globally and locally. The application of isotope techniques for these purposes occupies an increasingly important position, especially in the last fifty years in which nuclear instrumentation has reached a high technological level [1, 2].

The use of isotopes in hydrology depends on the nature of the problem according to which the classification can be made: tests related to the characteristics of surface watercourses (determination of flow, leakage of reservoirs and canals), examination of connections between groundwater and rainwater, thermal water testing, tests in sedimentology (determination of sediment formation rate), testing in limnology (determination of water measurement dynamics in accumulation), tests in hydrometeorology (determination of the formation zone of wet waves, and testing in glaciology (in the formation and transport of glaciers) [3, 4].

The use of isotopes in hydrology can be divided into two fields as: surface and groundwater can be labeled with artificial radioisotopes or substances that can later be activated (using activation analysis) and waters in their natural state have stable and radioactive isotopes that can also serve for labeling. The second solution is more expedient since the tracers are expected to follow the flow of water as completely as possible, and that they can be easily and accurately detected in low concentrations [5-7].

Insertion of radioactive tracers into groundwater is done by means of special wells or through natural cracks, and their detection is based on emitted radiation at points that can be wells or sources. The main disadvantage of this method is the possibility of loss of radioactive tracers by adsorption during flow through underground media.

In order to increase the efficiency of these methods, several radioactive tracers are used together, especially if different underground flows are tested, in partially coherent areas, under combined conditions.

Of all the isotopes that occur in nature, deuterium, oxygen ^{18}O, tritium and carbon ^{14}C are of particular interest for hydrological tests. Deuterium and oxygen ^{18}O are stable isotopes, found in water according to the proportionality determined by their abundance and deviation of physical properties in relation to the basic properties of hydrogen and oxygen ^{16}O. The third interesting isotope, tritium, is created in the atmosphere by cosmic radiation. Since it is radioactive, its creation was, until recently, balanced by decay. In recent times, human activities in the field of nuclear

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physics have significantly disturbed this balance. Carbon ^{14}C is also produced by cosmic radiation in the upper atmosphere. By oxidation, it remains for the most part in the atmosphere, and one part passes into the hydrological cycle via calcium carbonate.

The aim of this paper is to analyze the water circulation in Šar mountains aquatorium, Serbia, which is rich with water in an otherwise waterless environment. Oxygen isotopes ^{18}O and tritium will be used to achieve this goal, and statistical mathematics methods will be used to quantify the results [8, 9].

DETERMINATION OF TRITIUM AND OXYGEN ^{18}O CONTENT IN HYDROLOGICAL POPULATIONS

Determination of tritium in hydrological populations

Experimental determination of tritium activity of a hydrological population consists of following phases: sampling, sample preparation for enrichment, sample enrichment, sample preparation for measuring, and measuring tritium activity in the sample [10].

When taking a sample, care should be taken to ensure that it is a faithful representative of the analyzed water. It should also be noted that, after taking, the sample must not be in contact with atmospheric moisture.

To measure the activity of tritium, in principle, all standard methods used in the measurement of ionizing radiation can be used, in which gas meters, nuclear emulsions, semiconductor meters, etc. are used as detectors [11].

The main problem with measuring tritium activity using any of the aforementioned methods is that tritium emits low-energy beta particles, so it must be introduced directly into the active zone of the detector. In most cases, the tritium activity of precipitation, surface and groundwater samples is measured using scintillation counters. For this purpose, the liquid sample is mixed with a scintillator [12, 13].

Sample enrichment is the most important part of the natural tritium determination process. Enrichment with a factor of 10 to 100 is usually performed. All standard methods for isotope enrichment can be applied [14].

The enriched samples are mixed with a suitable scintillator (about 9 mL and 14 mL scintillator). It is important that the sample is retained in the mixture unchanged for more than 10 days, and also, a minimum level of background radiation should be obtained during the measurement.

The applied scintillation counter must have known linearity within the limits of measurement and should be stable and well protected from background radiation.

Determination of ^{18}O oxygen in hydrological populations

Samples are taken from the place where it is necessary to measure oxygen ^{18}O (river water, precipitation, groundwater). When taking samples for these measurements, bottles that are filled to the top with water and tightly closed to avoid isotopic exchange with atmospheric moisture, are usually used.

Content fluctuation in the oxygen ^{18}O isotope can only be accurately measured using a mass spectrometer. In order to avoid the memory effect of the mass spectrometer, it is necessary to convert the sample to a gaseous state. There are various techniques for converting aqueous samples to a gaseous state. In this paper, the procedure of bringing the sample into equilibrium with standard CO_2 is applied according to the relation [15]

$$CO_2^{16} \quad H_2O^{18} \qquad CO^{16}O^{18} \quad H_2O^{16} \qquad (1)$$

where the fluctuation of oxygen ^{18}O in $CO^{16}O^{18}$ according to standard CO_2 is measured after equilibration. To determine this fluctuation, the water sample is reduced over uranium to hydrogen, which is further measured.

The oxygen ^{18}O concentration was analyzed by mass spectrometer WG-MICROMASS designed exclusively for this type of measurement to yield isotopic fluctuations in relation to the standards applied.

MIXED STATISTICAL DISTRIBUTIONS

Analysis of the connection between precipitation and surface waters in an aquatorium, such as Šar mountains aquatorium, Serbia, using tritium and oxygen ^{18}O implies the application of statistical mathematics methods. Since these are water samples that have different histories, and since tritium is an unstable nucleon, statistical processing of the measurement results must be based on mixed distributions and the law of increasing probability. The resulting distribution of samples formed from precipitation, groundwater and surface water should be of the additive type, but the instability of tritium leads to the fact that these distributions can be of the multiplicative type. This includes the application of the model described by the Weibull distribution, which is based on the multiplication expression when calculating the probabilities. Therefore, multiplicative mixed distributions are treated as the application of the law of increasing probability [16, 17].

Additive and multiplicative mixed distributions are very difficult to process mathematically due to the large number of parameters. For this reason, empirical methods are needed that can be applied through some computer software, or simply graphically using the appropriate probability paper. These graphical methods

are based on the empirical density function, which are deductively divided into density functions of a special type of distribution so that the initial densities are obtained by superimposing the latter. The mitigating circumstance is, for example, the fact that when normal distribution probability paper is used, the graphs of the empirical functions are almost normally distributed if the component functions are also normally distributed. These real graphs should be the starting point in the graphical approximation [18, 19].

The law of increasing probability (which includes decrease) from the statistical point of view represents practical application of the law of multiplication of independent probabilities, P(A B) = P(A)P(B). Of course, the independence of individual probabilities that occur in parallel in relation to space and time, or consecutively, is implied.

For statistical processing of the increase or decrease of an entity in relation to space and time, a 4-D increase factor n is introduced, which represents the relationship between the increased (V_n, T_n) and reduced system (V_1,T_1)

$$n \quad \frac{V_n T_n}{V_1 T_1} \tag{2}$$

when some volume V_1, with the probability p_1 of the presence of the tracer, increases by n times then there is an increase in the probability of the occurrence of the tracer p_n, which is determined by the expression

$$1 \quad p_n \quad (1 \quad p_1)_1 (1 \quad p_1)_2 \ldots (1 \quad p_1)_n \tag{3}$$

where complementary events are taken into account. If it is assumed that the individual volumes are identical then eq. (3) passes into

$$p_n \quad 1 \quad (1 \quad p_1)^n \tag{4}$$

If instead of discrete probabilities the entire distribution functions $F_1(x)$ and $F_n(x)$ are taken into account then

$$F_n(x) \quad 1 \quad [1 \quad F_1(x)]^n \tag{5}$$

The enlargement now comes down to assuming an identically distributed variable for n elements considered by the number of tracers per sample unit [20-22].

In this case, eq. (5) turns into eq. (6)

$$F_n(x) \quad 1 \quad \prod_{i \ 1}^{n} [1 \quad F_{1i}(x)] \tag{6}$$

Equation (6) is a general expression independent of the statistical distribution of a stochastic sample of random variables, since the group of observed random variables is subject to other statistical distributions that give other expressions of the increasing probability based on the general eq. (6). The affiliation of a random variable to a specific statistical distribution stems from its nature, which can only be determined experimentally. At the end, the experimental results should be tested for belonging to those distributions that arise from the observed problem. To observe the distribution of natural nuclides of water components as tracers, minimum value type distributions and normal distribution are imposed. For this reason, in this paper we will consider the exponential distribution, the two-parameter and three-parameter distribution, and the normal distribution [23-25].

EXPERIMENT

The experimental procedure consisted of taking two identical samples 50 times in 0.5 dL, 1 dL, 2 dL, 5 dL, 7 dL, and 10 dL plastic bottles. Plastic bottles have been cleaned of memory effect, filled to the top and sealed in water (to avoid contact with atmospheric air). Sampling took one year at intervals of about 25 days (depending on the occurrence of precipitation).

After sampling, they were immediately measured for tritium and oxygen ^{18}O concentrations. The samples were first cleaned of suspicious results. After that, the samples were tested for belonging to a single statistical variable using the U-test.

Table 1 shows the dependence of the variation coefficient on enlargement factor for the exponential distribution, two-parameter Weibull distribution, three-parameter Weibull distribution and for the normal distribution.

Figure 1 shows the dependence of the variation coefficient from tab. 1 together with the experimentally obtained points.

From fig. 1 it can be seen that the concentration of tritium in all distributions belongs to the two-parameter Weibull distribution of the multiplicative type, and the oxygen ^{18}O concentration in all distributions belongs to the three-parameter Weibull distribution of the three-parameter type. It was also observed that the concentration of tritium in groundwater samples belongs to the two-parameter Weibull distribution of the additive type.

Based on the results shown in fig. 1, it was possible to express equations for the dependence of distribution functions of the oxygen ^{18}O and tritium in the tested samples [26].

In the case of oxygen ^{18}O, it was shown that the dependence curve belongs to the three-parameter Weibull distribution. If the Weibull distribution is taken for identical initial distributions

$$F_1(x) \quad 1 \quad \exp\{ \ [(x \quad x_0)/\eta_1]^{\delta} \} \tag{7}$$

and replace in the basic equation of the law of increasing probability, it follows that [27]

$$F_n(x) \quad 1 \quad \exp \quad \frac{x \quad x_0}{\eta_1}^{\delta} \quad n \tag{8}$$

Table 1. Dependence of variation coefficient on enlargement factor

	Distribution	Conversion from $\frac{s}{\bar{x}}_1$ to $\frac{s}{\bar{x}}_n$	Remarks
(1)	Weibull distribution (2 parameters); $x_0 = 0$	$\frac{s}{\bar{x}}_n = \frac{s}{\bar{x}}_1$	Variation coefficient is not dependent on enlargement factor
(2)	Weibull distribution (3 parameters)	$\frac{s}{\bar{x}}_n = \frac{s}{\bar{x}}_1 \frac{1}{1 \frac{x_0}{\bar{x}}(\sqrt[\delta]{n} 1)}$	Case (1) applies to $x_0 = 0$, x_0, δ, parameters Weibull distribution
(3)	Double exponential distribution	$\frac{s}{\bar{x}}_n = \frac{s}{\bar{x}}_1 \frac{1}{1 \frac{s}{\bar{x}}_1 \frac{\sqrt{6}}{\pi} \ln n}$	–
(4)	Normal distribution	$\frac{s}{\bar{x}}_n = \frac{s}{\bar{x}}_1 \frac{k_{1n}}{1 \frac{s}{\bar{x}}_1 k_{2n}}$	$k_{1n} \frac{1}{2}(\sqrt[\lambda n]{0.84} \sqrt[\lambda n]{0.16}$ $k_{2n} \frac{1}{2}(\sqrt[\lambda n]{0.84} \sqrt[\lambda n]{0.16}$ $\sqrt[\lambda n]{0.84}$ and $\sqrt[\lambda n]{0.16}$ are the quintiles of order $\sqrt[n]{0.84}$ and $\sqrt[n]{0.16}$ of an $N(0;1)$

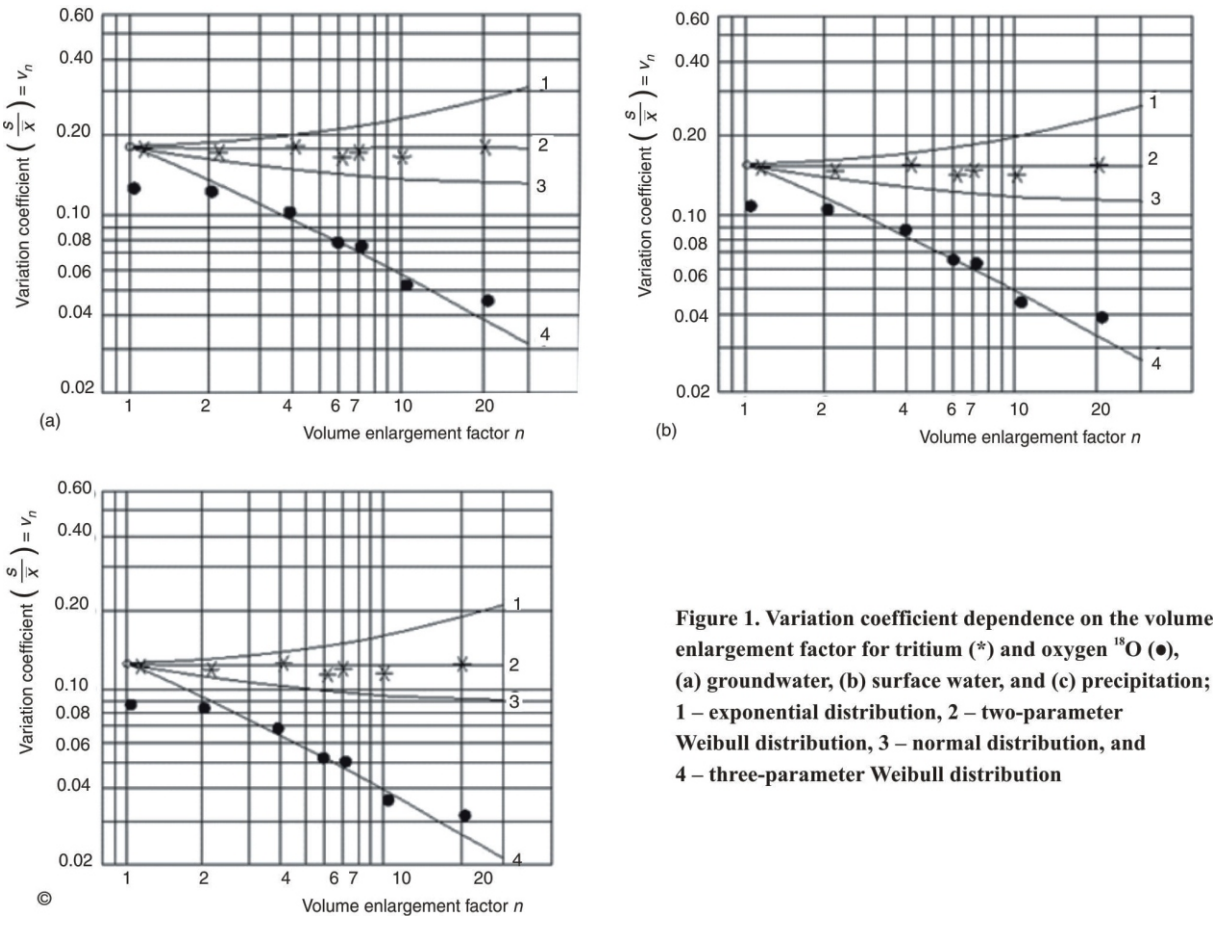

Figure 1. Variation coefficient dependence on the volume enlargement factor for tritium (*) and oxygen ^{18}O (•), (a) groundwater, (b) surface water, and (c) precipitation; 1 – exponential distribution, 2 – two-parameter Weibull distribution, 3 – normal distribution, and 4 – three-parameter Weibull distribution

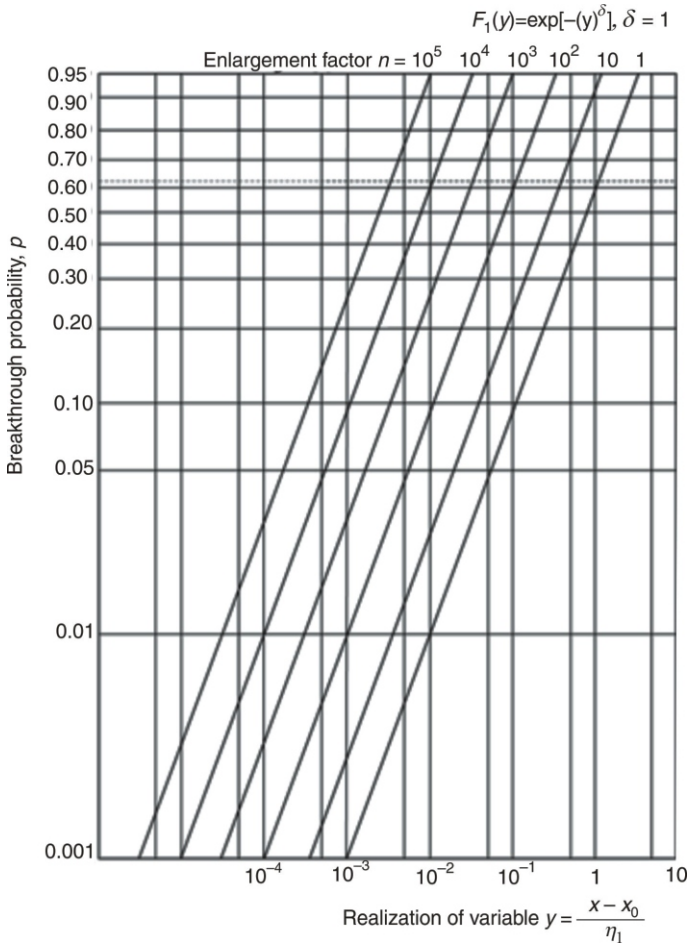

Figure 2. The law of increasing probability for the Weibull distribution, presented on probability paper (Weibull paper)

where η_1 , x_0 , δ, are the parameters of the initial distribution. In the case of factor n increase, only the 63 % quintile changes to

$$\eta_n \quad (x_{nm} \quad x_0)_{63} \quad \eta_1 n^{1/\delta} \quad (x_{1m} \quad x_0)_{63} n^{1/\delta} \quad (9)$$

while the initial value x_0 and the Weibull exponent, δ, remain the same. On probability paper, the distribution functions obtained from the reduced Weibull distribution ($\eta_1 = 1, x_0 = 0$, from tab. 1) are parallel to the initial distribution $F_1(x)$, fig. 2. The change in mode when there is enlargement, eq. (9), is shown in fig. 3 in relative form.

In the case of a parallel connection of n elements with a variable that is subject to the Weibull distribution, but with different parameters η_i, x_{0i} and $_i$ then, analogous to eq. (6), the general law of increasing probability gives

$$F_n(x) \quad 1 \quad \exp \quad \prod_{i \quad 1}^{n} \exp \left(\frac{x \quad x_{0i}}{\gamma_{1i}} \right)^{\delta_i} \quad (10)$$

In this case, as in the case of the double-exponential distribution, the type of distribution is lost.

Based on aforementioned, it is possible to perform simpler expression for standard deviation

$$\sigma_n \quad \frac{\sigma_1}{n^k} \quad (11)$$

and density mean value of the oxygen ^{18}O depending on basic sample enlargement by n times

$$n_n \quad n_1 \quad L(k) \left| 1 \quad 1/n^k \right| \sigma_1 \quad (12)$$

where $L(k) \quad \left| \Gamma(1 \quad 2k) \quad \Gamma^2(1 \quad k) \right|^{12} \Gamma(1 \quad k)$ and $\Gamma(k)$ is gamma function.

Based on eqs. (11) and (12), the increase in tritium concentration can be calculated without taking into account the radioactive decay. If the radioactive decay of tritium is taken into account (which is especially true for old groundwater samples) then follows ($\delta \quad 2$)

$$F_n(n_d) \quad 1 \quad \exp \quad \left(\frac{n_d}{n_{d63}} \right)^{\delta} \quad \frac{2\pi l V_i^2 \quad \left(\frac{V_a}{V_i} \right)^{2 \quad \delta} \quad 1}{(2 \quad \delta) V_1} \quad (13)$$

where n is

$$n \quad \frac{2\pi l V_i^2 \quad \left(\frac{V_a}{V_i} \right)^{2 \quad \delta} \quad 1}{V_1 (2 \quad \delta)} \quad (14)$$

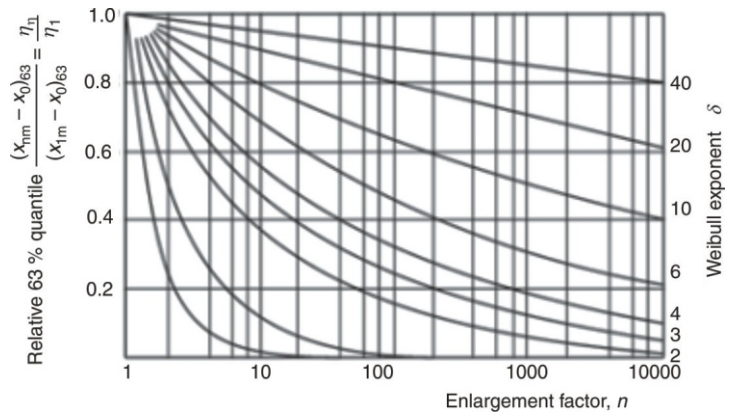

Figure 3. Parameter η_n $(x_{nm}$ $x_0)_{63}$ of Weibull distribution as a function of the enlargement factor n

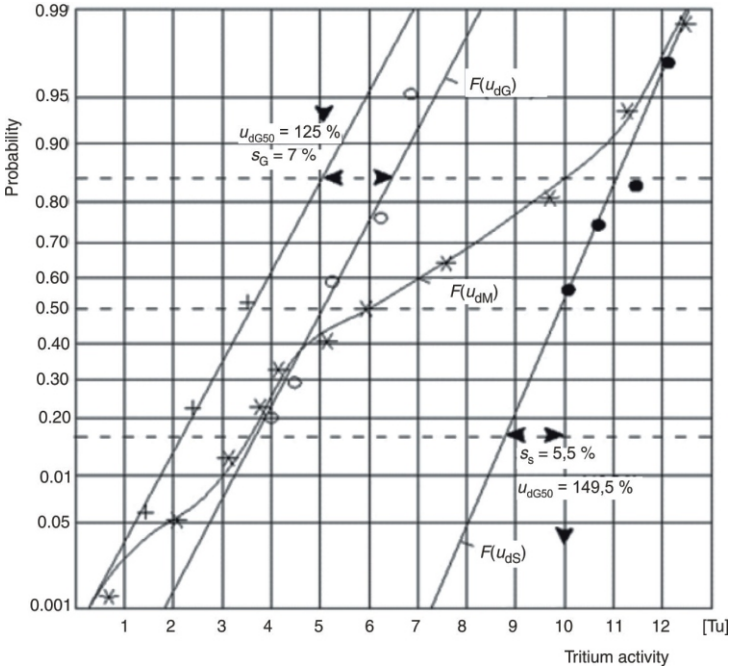

Figure 4. Tritium activity in a mixture of groundwater (+), surface water (○) and precipitation (●); resultant activity (*)

By applying the two-parameter Weibull distribution, the type of distribution is preserved and the 63 % quantile changes according to

$$n_{d63N} \quad n_{d63} \frac{V^1(2 \quad \delta)}{2\pi l V_i^2 \left(\frac{V_a}{V_i}\right)^{2\ \delta} \quad 1}^{1/\delta} \tag{15}$$

In order to verify the derived expressions, a significantly increased sample (100 L) of water from the lower parts of the Šar mountains was taken, where all the tested waters (precipitation, groundwater and surface water) were mixed and analyzed according to the expected results based on water measurements and expressions.

$$\frac{n_{d63_1}}{n_{d63_2}} \frac{\ln \frac{V_{a1}}{V_{i1}}}{\ln \frac{V_{a2}}{V_{i2}}} \frac{V_{i2}}{V_{i1}}^{(2/\delta)\ 1} \frac{\left(\frac{V_{a2}}{V_{i2}}\right)^{2\ \delta} \quad 1}{\left(\frac{V_{a1}}{V_{i1}}\right)^{2\ \delta} \quad 1}^{1/\delta} \tag{16}$$

With regard to the mathematical procedure, it should also be noted that the volume effect must be calculated using a physically appropriate selected variable. The result can be transformed at any time to express the desired variable, using known links to other variables.

Figure 4 shows the concentrations of tritium (Tu unit is defined as the ratio of one tritium atom to 10^{18} hydrogen atoms) in Jažinačko Lake. By statistical analysis, using the U-test revealed the existence of

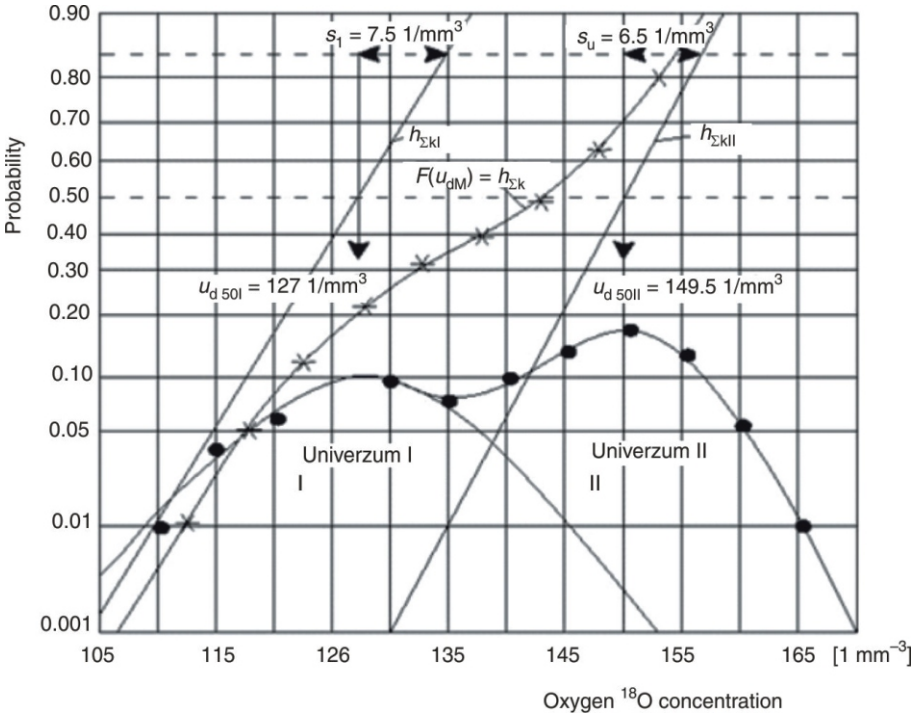

Figure 5. Oxygen ^{18}O concentration in a mixture of groundwater and surface water (●)

three statistical samples that can be clearly divided into groundwater generated before nuclear activities, *i.e.* before the creation of nuclear reactors, atomic bombs, *etc*. The group of random variables includes precipitation waters and it can be roughly estimated that most of the precipitation was in the summer period when the processes of nucleosynthesis in the atmosphere are much more active, *i. e*. experiencing a peak.

Figure 5 shows the oxygen ^{18}O concentration in Durlov Creek with clearly separated two areas (universes). The universe 1 belongs to the old groundwater in which the percentage of oxygen ^{18}O is approximately equal to its natural abundance. The universe 2 belongs to precipitation waters with increased concentration of oxygen ^{18}O due to the scientific research of human activity started sixties.

Figure 6 shows the activity of tritium in groundwater that has little or no contact with surface water. The diagram shows the low activity of tritium, which is characteristic of the prenuclear era. It can be seen the existence of a slight peak that is interpreted as a contact with surface water. The theoretical curve is in perfect agreement with eq. (16) thus confirming the theoretical analysis.

Because of the mathematical difficulties associated with mixed distributions, they should only be used when physically essential, *i.e*. when the physical model of the random process being investigated, produces a mixed distribution. One should on no account interpret an empirically – found relationship from the outset as a mixed distribution without such a model. No doubt, many relationships in nature and technology intrinsically follow mixed distributions, but in the end, in the areas that matter, it is usually individual influences that dominate, so that mixed components can

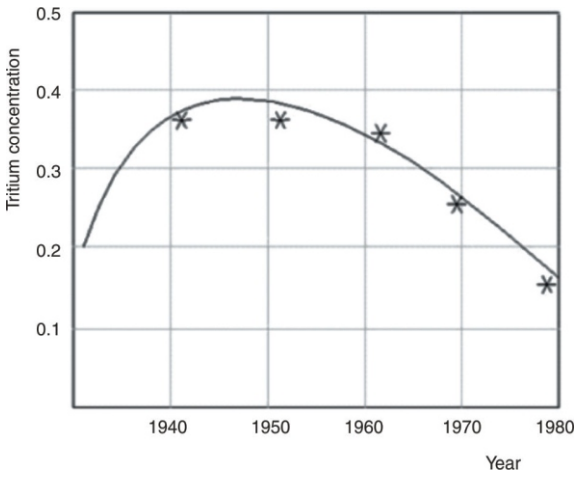

Figure 6. Groundwater concentration in samples taken from 1940 to 1980 and the corresponding curve according to eq. (16)

be ingnored. For example, when applying the law of increasing probability to the mixed distribution, it would be sufficient just to consider the normal distribution associated with universe 1.

CONCLUSION

The analysis of natural tracers in the composition of surface water, groundwater, and precipitation waters can fully establish the past, present and future of an aquatorium. The great advantage of this method is in the chronologically accurate prediction of the occurrence in the observed aquatorium. It is also possible

to predict the quantities of water of each origin and thus predict the possibility of their application in industry and water supply for the population. It should be especially emphasized that natural isotopes of helium and hydrogen are constituents of water (with a precisely determined abundance) and thus, their hydrodynamics are not affected by anything.

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AUTHORS' CONTRIBUTIONS

N. Z. Stanojević gave the idea for the experiment, which was carried out by J. V. Djokić and D. P. Nikezić. All authors analyzed the results and participated in preparation of the final version of the manuscript under supervision and guidelines of P. V. Osmokrović.

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ПРИМЕНА СЛОЖЕНИХ СТАТИСТИЧКИХ РАСПОДЕЛА И ПРИРОДНИХ ИЗОТОПА ВОДОНИКА И КИСЕОНИКА ЗА ПРОЦЕНУ ПОРЕКЛА ВОДА У АКВАТОРИЈУМУ ПЛАНИНЕ ШАРЕ

У раду се разматра могућност примене природних изотопа воде, трицијума и кисеоника ^{18}O, за анализу повезаности подземних, надземних и падавинских вода. Анализа омогућава и одређивање старости подземних вода издвојених из циклуса кружења воде у природи. На основу ових метода могуће је поуздано, хронолошки прецизно, одредити могућност индустријске примене и примене за људску потребу вода тог акваторијума.

Кључне речи: акваторијум планине Шаре, трицијум, кисеоник ^{18}O