

Research Article

Open Access

Marija M. Janković*, Nataša B. Sarap, Gordana K. Pantelić, Dragana J. Todorović,

Comparison of two different methods for gross alpha and beta activity determination in water samples

Abstract: Measurement of the gross alpha and beta activity concentrations in various matrices is suitable as a preliminary screening procedure to determine whether further analysis related to specific radionuclide is necessary. In Serbia, according to current regulations, radioactivity concentrations in drinking water for gross alpha and gross beta should be < 0.5 and < 1.0 Bq L⁻¹, respectively. The generally accepted methods for gross alpha and beta activity analysis of drinking water in different countries are: EPA 900.0 and ISO methods (ISO 9696, water quality – measurement of gross alpha activity in non-saline water – thick source method, and ISO 9697, water quality – measurement of gross beta activity in non-saline water). This paper describes comparison of preparations and measurement gross alpha and beta activity in bottled mineral waters as well as in tap waters in Serbia using two methods: EPA 900.0 with and without ash, and ISO methods. Concentration of total dissolved solids (TDS) in investigated water samples was determined. Based on obtained TDS, certain volume was evaporated. The instrumentation used to count the gross alpha and gross beta activities for both methods is α/β low level proportional counter Thermo Eberline FHT 770 T.

Keywords: natural radioactivity, gross alpha and gross beta activity, bottled mineral water, tap water.

DOI: 10.1515/chem-2015-0081

received February 28, 2014; accepted July 13, 2014.

*Corresponding author: **Marija M. Janković:** University of Belgrade, Institute of Nuclear Science Vinča, Radiation and Environmental Protection Department, 11001 Belgrade, Serbia, E-mail: marijam@vinca.rs

Nataša B. Sarap, Gordana K. Pantelić, Dragana J. Todorović: University of Belgrade, Institute of Nuclear Science Vinča, Radiation and Environmental Protection Department, 11001 Belgrade, Serbia

1 Introduction

Gross alpha/beta measurement is one of the simplest radioanalytical procedures which are applied widely as a screening technique in the field of radioecology, environmental monitoring and industrial applications. This procedure has been developed to determine whether radionuclide specific analysis is required to further characterize the water. Regular measurements of gross alpha and gross beta activity in water may be invaluable for early detection of radioactive contamination and indicate the need for supplemental data on concentrations of more hazardous radionuclides.

Water is the most important and widespread natural resource, and it is an essential compound taken in and excreted by all living organisms. The quality of water has a profound impact on human lives because it is so commonly consumed and used by households. Therefore, to minimize health hazards, the quality of water should be constantly monitored and analyzed with the aim of acquiring reliable information about the level and trends of water pollution [1].

In the laboratory for radiation protection and environmental protection in the Institute of Nuclear Sciences Vinča in Serbia, drinking, mineral, tap, well water, surface water as well as groundwater are assayed for the gross alpha and gross beta activity. Generally, in many laboratories around the world the gross alpha and beta activity in waters are investigated in order to determine potential health hazards from natural radionuclides in consuming water [2]. Permissible activity concentrations in drinking water are recommended in Guidelines for Drinking Water Quality (World Health Organization) [3]. Contribution to the total alpha activity comes primarily from Ra, Th, U, Rn and its decay product, while contribution to the total beta activity comes from ⁴⁰K, natural or artificial tritium, artificial ⁹⁰Sr and artificial ¹³⁷Cs [4]. The most frequently applied methods for the determination of gross alpha and beta activities in drinking water are

EPA method [5-8] and ISO methods [9-12]. These methods involve evaporation of certain volume of water to dryness, ashing at 450°C and counting the residue deposited on a planchet by a gas proportional detector.

On the other hand, other methods are also developed such as method given in references [13,14]. In addition to the gas proportional detectors, determination of gross alpha and beta activity can be conducted on a liquid scintillation counter [11,13-20] as well as combined gamma-alpha spectrometry technique [2].

As already mentioned, determination of gross alpha and beta activity is screening method. If the obtained value for gross alpha activity in drinking water samples exceeds the allowed value of 0.5 Bq L⁻¹ [21,22] and the obtained value for gross beta activity exceeds the allowed value of 1 Bq L⁻¹ [21,22], then other methods such as gamma spectrometry or determination of ⁹⁰Sr by analytic method, or determination of tritium by liquid scintillation counter must be applied. If the obtained values are less than above mentioned values, water can be used for drinking [23].

The objective of this work is to apply different methods for drinking water sample preparation and to see where better results are obtained.

2 Experimental Procedure

2.1 Methods

2.1.1 EPA (Method 900.0) gross alpha and beta radioactivity in drinking water:

According EPA method, certain volume is calculate which is needed to be evaporated, in order to after ashing at 450°C residue should be 0.1A mg (where A is planchet area in mm²). Total dissolved solids (TDS) must be determined at first. The certain volume of water sample then evaporated under infrared lamp to dryness. For a 6 cm diameter counting planchet (28.26 cm²) (planchet used in our laboratory), an aliquot containing 130 mg of dissolved solids would be the maximum aliquot size for that sample which should be evaporated and counted for gross alpha activity. For gross beta activity, for solids density thickness of 10 mg cm⁻², 260 mg of dissolved solids would be the maximum aliquot size for that sample which should be evaporated and counted for gross beta activity. We used two EPA methods: in the first 3 L of waters was evaporated and ash at 450°C. Residue are transferred in planchet (130 mg) and measured. In the second, certain volume was evaporated near to dryness and residue

are transferred in planchet and evaporated to dryness. The sample residue is than dried in a drying oven at 105°C for at least 2 hours; cool in a desiccator; weigh; and count. Samples may be counted for beta activity immediately after preparation, but alpha counting should be delayed at least 72 hours until equilibrium has occurred.

2.1.2. ISO 9696 Water quality – measurement of gross alpha activity in non-saline water – thick source method; ISO 9697 Water quality – measurement of gross beta activity in non-saline water

According ISO methods [9,10], certain volume is calculate which is needed to be evaporated, in order to after ashing at 450°C residue should be 0.1A mg (where A is planchet area in mm²). The largest sample aliquot that should be counted for gross alpha and beta activity is the same as in EPA procedure described above. Generally, method includes evaporation of certain volume to the dry residue, translation in the sulphate form and ashing at 450°C for an 24 hours. The residues were transferred quantitatively to a stainless-steel planchet and count. Measurements were performed in the same way as in EPA method.

2.1.3. Measurement of gross alpha and beta activity

Detector used for gross alpha and beta activity measurement in water samples were α/β low level proportional counter Thermo Eberline FHT 770 T. Calibration of detector and calculation of activity was performed in the same manner as described in reference [7].

3 Results and Discussion

pH, conductivity, dry residue and main dissolved ions concentrations in five investigated bottled water samples (Rosa, Aqua Una, Premia, Vrnjci and Minaqua bottled mineral waters produced in Serbia) are shown in Table 1. Concentration of minerals in all water samples (dried at 180°C) ranged between 56 and 1184 mg L⁻¹. Three investigated waters belonged to the medium-mineral class (residue > 500 mg L⁻¹) and the other two – to the low-mineral class (residue 50–500 mg L⁻¹). The pH value for Minaqua water was the lowest compared to other investigated samples. All waters were carbonated (HCO₃⁻). Content of cation was in a large range, in some waters Na and K ions prevailed – Vrnjci and Minaqua (water contains large amount of sodium, if Na⁺ concentration exceed 200 mg L⁻¹ [24,25]). Premia contained large amount of Ca ions (water contains large amount of calcium if Ca²⁺

Table 1: Chemical parameters of 5 brands of bottled mineral water samples (mg L⁻¹).

Brand	Rosa	Aqua Una	Premia	Vrnjci	Minaqua
pH	7.93	7.08	6.42	7.62	5.93
TDS	56	324	848.5	939	1184
Conductivity / μS cm ⁻¹	9	56			1755
Ca ²⁺	10	60.1	232.4	68	31.8
K ⁺	< 1	2.7	5.6	29.5	4
Mg ²⁺	0.91	32.8	18.2	51.5	20.7
Na ⁺	2.7	16.7	56	203.7	397.7
Cl ⁻	< 1	1.85	14.4	18	306.4
Fe ^{2+/3+}		< 0.05			
F ⁻		0.14		1.33	
I ⁻					0.94
SO ₄ ²⁻	5.4	13.4	11.3	38.10	
HCO ₃ ⁻	42.7	402.6	988	994.30	757
SiO ₂	13.7				
NO ₃ ⁻	1.3				
Content of CO ₂ (min)			3000	3000	3000

concentration is higher than 150 mg L⁻¹ [24,25]), Vrnjci was rich in Mg ions (water contains large amount of magnesium if Mg²⁺ concentration exceed 50 mg L⁻¹ [24,25]), Minaqua – in Cl ions (water contains large amount of chloride if concentration of Cl⁻ is higher than 200 mg of chloride per liter [24,25]), and Vrnjci – in F ions (water contains large amount of fluoride if F⁻ exceed 1.0 mg L⁻¹ [24,25]). On the other hand, Rosa and Aqua Una had low sodium content, less than 20 mg L⁻¹ [24,25]. High conductivity value for Minaqua confirms its natural mineralization. Values presented in Table 1 were obtained for samples taken directly from bottles. Total dissolved solids (TDS) shown in Table 1 were obtained after sample evaporation and subsequent drying the remaining residue at 180°C (for bottled water). Since, the required amount of water to be evaporated for the determination of the gross alpha and beta activity is dependent on TDS (methods EPA and ISO), this parameter was determined once more using slightly different procedure. This second TDS measurement was based on the weight of the solid residue remained after evaporation of sample and subsequent drying at 105°C. After evaporation of sample (40 mL) residue was dried for at least 2 h in an oven at 105°C, cooled in a desiccator,

and weighted. Cycles of drying, cooling, desiccating, and weighting were repeated until a constant weight was obtained or until weight change was less than 4% of the previous weight or 0.5 mg. For counting gross beta activity in a water sample the TDS value is not as limiting as for gross alpha activity because beta particles are not stopped in solids as easily as alpha particles are. Obtained results are presented in Table 2. As can be seen these results are in good agreement with TDS obtained from bottle (for bottled waters), except for Rosa water, where TDS obtained at 105°C are higher than the result obtained at 180°C (Table 1).

Together with bottled mineral waters, tap water from cities in Serbia: Belgrade (Vinča), Topola, Batrovci, Požarevac and Lučani, was prepared for determination of gross alpha and beta activity using two different methods. In these samples pH and conductivity were determined initially. The pH was measured by InoLab pH meter WTW using glass electrode SenTix 81. Calibration of the instrument was carried out by 4, 7 and 10 pH standard solutions. The conductivity of the samples was measured by Conductometer InoLab WTW Cond7110 at 20°C. pH values: Belgrade 7.39, Topola 7.55, Batrovci 8.05, Požarevac 7.00 and Lučani 7.69. The waters proved to be weakly basic except of Požarevac which was neutral. Conductivity of these waters were: Belgrade 419 μS cm⁻¹, Topola 455 μS cm⁻¹, Batrovci 615 μS cm⁻¹, Požarevac 960 μS cm⁻¹ and Lučani 342 μS cm⁻¹. Differences in conductivity values are related to the presence of different amounts of mineral salts and their origin. Conductivity values were in the range of 9–1755 μS cm⁻¹, both for bottled mineral water and for tap water. According to Regulation [24], recommended value for conductivity should reach up to 2500 μS cm⁻¹.

Table 2 presents results of gross alpha and beta activity concentrations for all analyzed samples obtained using following methods:

- E1 – EPA 1, a modified EPA method used in Radiation and Environmental Protection Department, Institute Vinca, University of Belgrade. Water sample of 3 L was evaporated and the remained residue was ashed at 450°C. It is essential to analyze as large sample aliquot as is needed to give reasonable counting times to meet the required sensitivities (detection limits), especially for samples with very low concentrations of radioactivity such as drinking water samples,
- E2 – EPA 2, where initial volume of a sample is a function of total dissolved solids. This method includes evaporation to dryness and weighting without ashing,

- I – ISO, initial volume of the sample is a function of total dissolved solids. This method includes evaporation to dryness, adding concentrated sulfuric acid and ashing at 450°C.

Results obtained both immediately after preparation and after 72 hours are presented.

What is important, for all water samples gross alpha and beta activity turned out to be lower than the recommended level (0.5 Bq L⁻¹ for gross alpha activity and 1 Bq L⁻¹ for gross beta activity [22]) regardless of which method was used, except Vrnjci water where gross beta activity was higher than 1 Bq L⁻¹ (methods E1 and E2). For the latter γ spectrometric analysis was performed (results are given below). If one of the values is higher than recommended, radionuclides have to be identified by alpha and/or gamma spectroscopy, and their individual activity concentrations need to be measured.

It can be noted that results obtained for gross alpha and beta activity using methods E1 and E2 are quite consistent. For gross beta activity higher compatibility exists for water of high mineral content. These conclusions refer to the bottled waters. For tap water samples, results obtained for gross beta activity using E2 method were significantly lower than when E1 method was applied. The exception was water from Požarevac (tap water with the highest dry residue), where gross beta activity obtained using E2 method was higher when the sample was measured immediately after preparation. Also, for tap water from Belgrade gross beta activity obtained using E2 method was higher than activity obtained using E1 method, when sample measured after 72 hours. For gross alpha activity, the results were typically below the MDC (Minimum Detectable Change). MDC is different from the method to method primarily because samples were not always measured using the same detector. For example MDC for gross alpha activity for water Minaqua was 0.310 Bq L⁻¹ for E1, 0.250 Bq L⁻¹ for E2 and 0.313 Bq L⁻¹ for I method (Table 2).

However, it should be noted that experience has shown that it is better to evaporate large quantities of water to concentrate the samples, especially when the activity is expected to be low. That is, in our case, using the method E1. On the other hand, there are differences between parameters measured for samples prepared using E1, E2 and I method. In fact, it is worth to be noted that at low mineral waters (Rosa and Aqua Una), gross alpha and beta activity determined by ISO method were lower than the activity obtained using EPA methods, while in waters of higher mineral content (Premia and Minaqua), gross alpha and beta activity obtained using ISO method

were higher than the results obtained using EPA methods. This is a consequence of sulfuric acid addition to the sample, since the addition of an acid to water of large mineral contents resulted in partial precipitation. Water samples during the drying and ashing processes may give a residue unsuitable for the radioactivity measurement because of its hygroscopic properties. For this reason, sulfuric acid was added. Alpha and beta activity measured for Vrnjci water using ISO method was lower than when EPA method was applied. For tap waters only gross beta activity obtained using ISO method was lower than the results obtained using EPA method. Gross alpha activities for all tap waters obtained by ISO method fall below the MDC.

Differences between results obtained immediately after preparation and after 72 hours are summarized in Table 2. Gross alpha activity measured after 72 hours was generally lower, except for Aqua Una using E1 method. On the other hand, gross beta activity was independent of equilibrium, but results obtained after 72 hours were lower as well, except for Minaqua when E1 method was applied and for tap water from Belgrade using E2 method.

To one sample of Minaqua bottled water (sample prepared using E1 method) the nitric acid was added to volatilize chlorides during sample evaporation. Such amount of nitric acid was used to bring the pH of the sample to the value below 2.0. This was done to prevent precipitation of the radionuclides and other metals that may be present in the sample and co-precipitate the radionuclides. Nitric acid was not added to the other sample of Minaqua water. Gross alpha and beta activity obtained for sample to which nitric acid was added was < 0.112 Bq L⁻¹ and 0.449 ± 0.070 Bq L⁻¹ respectively. Gross alpha activity for Minaqua water without nitric acid obtained using E1 method was < 0.112 Bq L⁻¹ and gross beta activity 0.490 ± 0.100 Bq L⁻¹. Determination of gross alpha and beta activities for this sample was not affected by pH of the sample. In opposite to bottled waters, all tap waters were acidified using nitric acid to pH 2 and held in the original container for minimum 16 h before analysis.

Figs. 1-3 present the dependence of gross beta activity of TDS for different methods. A good correlation was found between gross beta activity and TDS ($r = 0.64$ for E1 method, $r = 0.71$ for E2 method and the highest linear correlation $r = 0.93$ was obtained for ISO method). These results show that beta activity concentrations are related to the dry residue of the samples, i.e. to the content of salts in the analyzed waters.

As shown above Vrnjci water has gross beta activity higher than the recommended level (1 Bq L⁻¹ [22]).

Table 2: Summarized data on TDS, gross alpha and gross beta activity concentrations in waters using different methods. The results are presented as the mean value of five measurements.

Brand	TDS (mg L ⁻¹)	E1		E2		I		
		Gross alpha activity (Bq L ⁻¹)	Gross beta activity (Bq L ⁻¹)	Gross alpha activity (Bq L ⁻¹)	Gross beta activity (Bq L ⁻¹)	Gross alpha activity (Bq L ⁻¹)	Gross beta activity (Bq L ⁻¹)	
Rosa (Vlasina)	78	1*	0.025 ± 0.005	0.084 ± 0.008	< 0.021	0.056 ± 0.010	0.013 ± 0.005	0.042 ± 0.008
		2	< 0.015	0.067 ± 0.007	< 0.021	0.052 ± 0.010	< 0.007	0.028 ± 0.007
Aqua Una (Arandelovac)	348	1	0.061 ± 0.020	0.205 ± 0.040	< 0.091	0.205 ± 0.040	< 0.059	0.177 ± 0.020
		2	0.083 ± 0.020	0.173 ± 0.030	0.067 ± 0.020	0.202 ± 0.030	< 0.059	0.169 ± 0.020
Premia (Neresnica)	910	1	< 0.212	0.421 ± 0.080	< 0.182	0.460 ± 0.080	< 0.258	0.726 ± 0.100
		2	< 0.199	0.357 ± 0.070	< 0.182	0.434 ± 0.080	< 0.258	0.676 ± 0.100
Vrnjci (Vrnjačka Banja)	923	1	0.206 ± 0.060	1.202 ± 0.100	< 0.214	1.160 ± 0.100	0.124 ± 0.070	0.924 ± 0.100
		2	< 0.240	1.072 ± 0.110	< 0.248	1.160 ± 0.100	0.110 ± 0.060	0.874 ± 0.100
Minaqua (Novi Sad)	1125	1	< 0.310	0.431 ± 0.100	< 0.310	0.426 ± 0.100	< 0.313	0.936 ± 0.200
		2	< 0.310	0.490 ± 0.100	< 0.250	< 0.388	< 0.313	0.916 ± 0.200
Topola	115	1	< 0.044	0.137 ± 0.036	< 0.021	0.060 ± 0.017	< 0.020	0.056 ± 0.010
		2	< 0.044	0.090 ± 0.032	< 0.026	0.033 ± 0.017	< 0.020	0.040 ± 0.010
Lučani	183	1	0.080 ± 0.026	0.223 ± 0.039	0.075 ± 0.024	0.148 ± 0.030	< 0.059	0.125 ± 0.025
		2	0.046 ± 0.020	0.161 ± 0.034	< 0.043	0.093 ± 0.025	< 0.059	0.114 ± 0.023
Belgrade	245	1	< 0.039	0.167 ± 0.036	< 0.079	0.161 ± 0.043	< 0.043	< 0.103
		2	< 0.038	0.143 ± 0.035	< 0.052	0.164 ± 0.040	< 0.043	< 0.099
Batrovci	370	1	< 0.060	0.162 ± 0.043	< 0.061	< 0.061	< 0.100	< 0.177
		2	< 0.082	< 0.100	< 0.041	< 0.058	< 0.091	< 0.173
Požarevac	668	1	< 0.104	< 0.126	< 0.129	0.213 ± 0.083	< 0.095	< 0.159
		2	0.074 ± 0.041	0.152 ± 0.061	< 0.240	< 0.187	< 0.101	< 0.179

1* measured immediately after preparation

2 measured after 72 hours

tap water

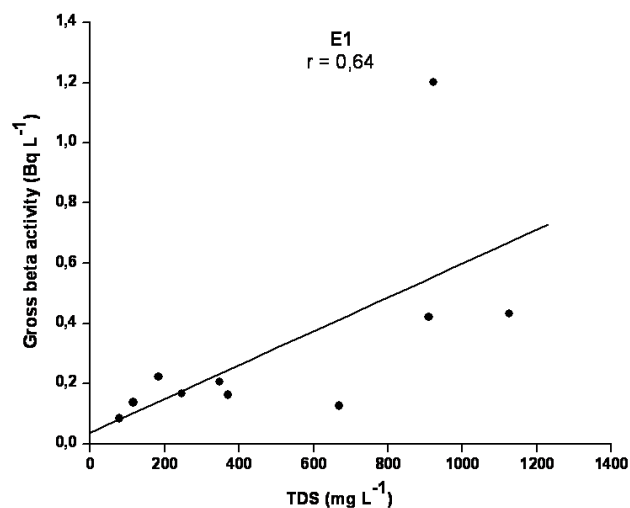
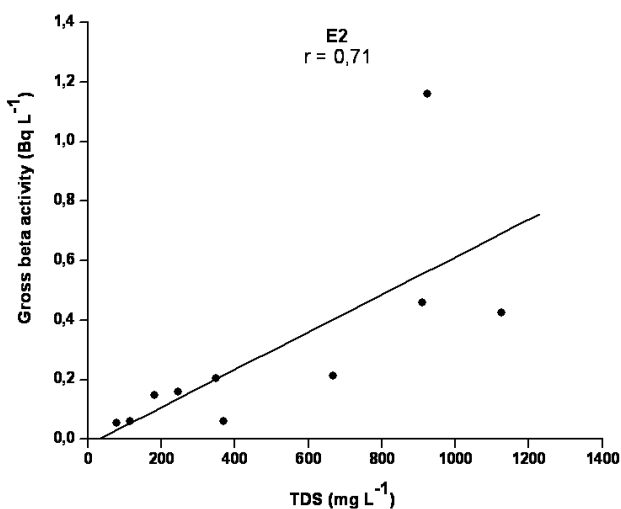
**Figure 1:** Correlation between gross beta activity and the total dissolved solids using E1 method.**Figure 2:** Correlation between gross beta activity and the total dissolved solids using E2 method.

Table 3: Results of participation in an intercomparison exercise.

	Water A	Gross beta activity (Bq L ⁻¹)	Water B	Gross beta activity (Bq L ⁻¹)	Water C	Gross beta activity (Bq L ⁻¹)
EC results	47.5 ± 22.8	309.8 ± 57.4	434.7 ± 56.6	190.4 ± 32.6	954.5 ± 77.3	1037.3 ± 83
LRM results	<120	413 ± 73	575 ± 60	821 ± 74	533 ± 39	1073 ± 77
TDS (EC) (mg L ⁻¹)	955		364		10.2	
TDS (LRM) (mg L ⁻¹)	1025		344		12.86	

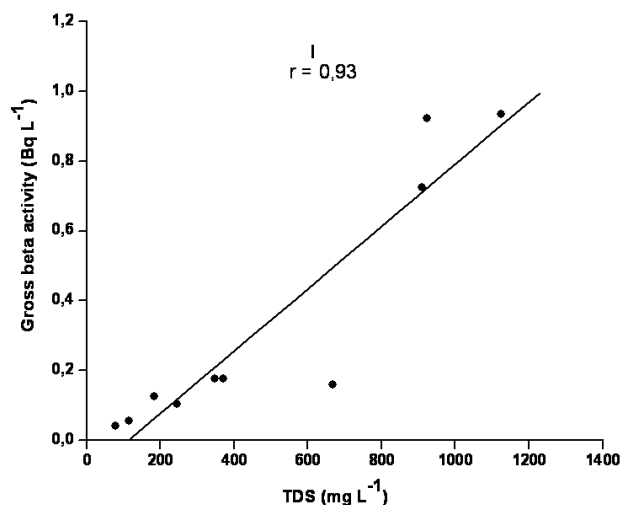


Figure 3: Correlation between gross beta activity and the total dissolved solids using I method.

The values obtained are: 1.202 Bq L⁻¹ using E1 method and 1.160 Bq L⁻¹ using E2 method. For gamma spectrometric analysis of this water 10 L of sample was evaporated to a small volume under infrared lamp. The residues were heated to dryness at 450°C [26] and transferred to a plastic box of 100 cm³. Gamma spectrometric measurements were performed using HPGe Canberra detectors with a relative efficiencies of 18%, 20% and 50%. Counting time interval was 60 000 s. Calibration of detectors for measurement of water samples was performed using secondary reference material in the plastic box of 100 cm³. Secondary reference material was obtained from the primary reference liquid radioactive material (Czech Metrological Institute, Prague) spiked with a series of radionuclides (²⁴¹Am, ¹⁰⁹Cd, ¹³⁹Ce, ⁵⁷Co, ⁶⁰Co, ²⁰³Hg, ⁸⁸Y, ¹¹³Sn, ⁸⁵Sr, ¹³⁷Cs, ²¹⁰Pb). The activity of ⁴⁰K was determined from its 1460 keV γ -energy. Radionuclide ⁴⁰K was detected in Vrnjci water and its concentration was 1.1 ± 0.1 Bq L⁻¹. Since the ⁴⁰K contributed to the total beta activity in 89%, we concluded that the gross beta activity originated mainly from ⁴⁰K. Moreover, presence of K⁺ ions

in Vrnjci water in an amount of 29.5 mg L⁻¹ (Table 1) might be the cause of increased beta activity. Concentration of natural radionuclides ²²⁶Ra, ²³²Th, ²³⁸U, ²¹⁰Pb and ²³⁵U in Vrnjci water were < 0.01, 0.020 ± 0.005, < 0.04, < 0.005 and < 0.002, respectively. Artificial radionuclide ¹³⁷Cs was < 0.001.

Radiation and Environmental Protection Department Institute Vinča (LRM Laboratory for Radiation Measurements) take part in intercomparison “EC interlaboratory comparison on gross alpha/beta activity measurement in drinking waters” organized by European Commission joint research centre in 2012. Determination of gross alpha and beta activity was performed in three drinking water samples A, B and C using E1 method. Water C was spiked with alpha and beta emitting isotopes. In all samples total dissolved solid content was determined. Preliminary results are given in Table 3. Results also published in [27].

4 Conclusions

Comparison of preparations and measurement of gross alpha and beta activity in five bottled mineral waters produced in Serbia and five tap water from different cities in Serbia using two different methods EPA and ISO were presented. Beside gross alpha and beta activity, total dissolved solids (TDS) in all water samples were determined. Based on obtained TDS, certain volume of water was evaporated. Results show that all waters have gross alpha and beta activity below than recommended level of 0.5 Bq L⁻¹ for gross alpha activity and below 1 Bq L⁻¹ for gross beta activity, regardless of which method was used, except Vrnjci water where gross beta activity was higher than 1 Bq L⁻¹ (E1 and E2 methods). For this water gamma spectrometric analysis was performed. It was concluded that higher gross beta activity in Vrnjci water originated mainly from ⁴⁰K, which concentration was

1.1 Bq L⁻¹. Linear correlation between gross beta activity in water samples obtained using E1, E2, I methods and TDS was examined and good correlation was found ($r = 0.64$ for method E1, $r = 0.71$ for method E2 and $r = 0.93$ for method I). Gross beta activity concentrations were related to the dry residue of the samples. The obtained results and practice show that EPA is a better method than ISO. It is necessary to apply the comparison of these described methods in a number of samples that include surface, groundwater and waste water.

Acknowledgement: The investigation was partially supported by The Ministry of Education, Sciences and Technological Development of Republic of Serbia, under Project III43009.

References

- [1] Cieszynska M., Wesolowski M., Bartoszewicz M., Michalska M., Assessment of river water quality in the South Baltic coast by multivariate techniques, *Cent. Eur. J. Chem.*, 2011, 9, 265-274.
- [2] Bonotto D.M., Bueno T.O., Tessari B.W., Silva A., The natural radioactivity in water by gross alpha and beta measurements, *Radiat. Measur.*, 2009, 44, 92-101.
- [3] WHO (World Health Organization), *Guidelines for Drinking Water Quality: Radiological Aspects*, 3rd edition, Geneva, 2004.
- [4] Semkow T.M., Parekh P.P., Principles of gross alpha and beta radioactivity detection in water, *Health Phys.*, 2001, 81, 567-574.
- [5] EPA (Environmental Protection Agency), *Prescribed Procedures for Measurement of Radioactivity in Drinking Water*, EPA-600/4-80-032; Method 900.0, 1980.
- [6] Turhan S., Ozcitak E., Taskin H., Varinlioglu A., Determination of natural radioactivity by gross alpha and beta measurements in ground water samples, *Water Research*, 2013, 47, 3103-3108.
- [7] Janković M.M., Todorović D.J., Todorović N.A., Nikolov J., Natural radionuclides in drinking waters in Serbia, *Appl. Radiat. Isot.*, 2012, 70, 2703-2710.
- [8] Janković M.M., Sarap N.B., Todorović D.J., Joksić J.D., Natural and artificial ⁹⁰Sr radionuclides in some carbonated mineral waters used in Serbia, *Nucl. Techn. Radiat. Protect.*, 2013, 28, 284-292.
- [9] ISO (International Standardization Organization) 9696, *Water Quality-Measurement of Gross Alpha Activity in Non-Saline Water-Thick Source Method*, Geneva, 2007.
- [10] ISO (International Standardization Organization) 9697, *Water Quality-Measurement of Gross Beta Activity in Non-Saline Water-Thick Source Method*, Geneva, 2008.
- [11] Kleinschmidt R.I., Gross alpha and beta activity analysis in water—a routine laboratory method using liquid scintillation analysis, *Appl. Radiat. Isot.*, 2004, 61, 333-338.
- [12] Jobbagy V., Kavasi N., Somlai J., Dombovari P., Gyongyosi C., Kovacs T., Gross alpha and beta activity concentrations in spring waters in Balaton Upland, Hungary, *Radiat. Measur.*, 2011, 46, 159-163.
- [13] Wisser S., Frenzel E., Dittmer E., Innovative procedure for the determination of gross-alpha/gross-beta activities in drinking water, *Appl. Radiat. Isot.*, 2006, 64, 368-372.
- [14] Happel S., Letessier P., Ensinger W., Eikenberg J.H., Thakkar A.H., Horwitz E.P., Gross alpha determination in drinking water using a highly specific resin and LSC, *Appl. Radiat. Isot.*, 2004, 61, 339-344.
- [15] Sanchez-Cabeza J.A., Pujol L., A rapid method for the simultaneous determination of gross alpha and beta activities in water samples using a low background liquid scintillation counter, *Health Phys.*, 1995, 68, 674-682.
- [16] Rusconi R., Forte M., Caresana M., Bellinzona S., Cazzaniga M.T., Sgorbati G., The evaluation of uncertainty in low-level LSC measurements of water samples, *Appl. Radiat. Isot.*, 2006, 64, 1124-1129.
- [17] Staffova P., Determination of Gross alpha and Beta activities in water samples by liquid scintillation counting, *Czechoslovak J. Phys.* 2006, 56, 299-305.
- [18] Todorovic N., Nikolov J., Tenjovic B., Bikit I., Veskovic M., Establishment of a method for measurement of gross alpha/beta activities in water from Vojvodina region, *Radiat. Measur.*, 2012, 47, 1053-1059.
- [19] Lin C., Wang J., Huang J., Yeh C., Yuan M., Chang B., Evaluating practicability of an LSC method for routine monitoring gross alpha and beta activities in water samples in Taiwan, *Appl. Radiat. Isot.*, 2012, 70, 1981-1984.
- [20] Fons J., Zapata-García D., Tent J., Laurado M., Simultaneous determination of gross alpha, gross beta and ²²⁶Ra in natural water by liquid scintillation counting, *J. Environ. Radioact.*, 2013, 125, 56-60.
- [21] WHO (World Health Organization), *Guidelines for Drinking Water Quality*, 3rd edition including the first addendum Geneva, 2006.
- [22] Official Gazette of the Republic of Serbia, Regulation on limits of radionuclide content in drinking water, foodstuffs, feeding stuffs, drugs, items of general use, building materials and other goods to be placed on the market, 86/11, Serbia, 2011.
- [23] Jobbagy V., Watjen U., Meresova J., Current status of gross alpha/beta activity analysis in water samples: a short overview of methods, *J. Radioanal. Nucl. Chem.*, 2010, 286, 393-399.
- [24] Official Gazette of the Republic of Serbia, Regulation on quality and other requirements for natural mineral water, spring water and bottled drinking water, 53/05, Serbia 2005.
- [25] Official Gazette of the Republic of Serbia, Regulation on the quality of natural mineral water, 37/2005.
- [26] IAEA, *Measurements of Radionuclides in Food and the Environment*, Technical Report Ser. No. 295. IAEA, Vienna, 1989.
- [27] Jobbagy V., Merešova J., Watjen U., Critical remarks on gross alpha/beta activity analysis in drinking waters: Conclusions from a European interlaboratory comparison, *Applied Radiation and Isotopes*, 2014, 87, 429-434.