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Drinking water purification in integrated system

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The drinking water resources of South-West Bulgaria and particularly in the region of Blagoevgrad are discussed. The main contaminants of tap water, spring water and ground water for drinking purposes are shown. Tap water is that which we get directly from the tap and it may or may not be suitable for drinking purposes. It is used for household purposes such as cleaning, cooking, gardening, laundry and more. The quality of drinking water must comply with the rules set by local municipal authorities. The national and EC requirements for the quality of drinking water are presented. The quality of drinking water is improved locally by filtration, reverse osmosis (RO) and UV disinfection. The applications of local water purification systems as a 7-stage system with RO and UV disinfection and a simple system with filtration, UV disinfection and pump are critically discussed. The 7-stage reverse osmosis system is a system that removes up to 99% of all harmful impurities from water: fluorine, pesticides, chlorine, chloramines, heavy metals, radioactive elements, microorganisms. After chemical analysis of water purification recommendations for a local improving quality of water by applications of advanced innovative water purification systems are presented.

Keywords: Drinking water, Filtration, Reverse osmosis, UV disinfection, Water purification systems

INTRODUCTION

Water is one of the most important natural resources on the Earth [1]. Although a large part of our planet (71%) is covered with water, only a small part can be used for drinking purposes (about 1%) [2]. Water consumption depends on various factors. The daily requirement of water for an adult male is 3 *l/day,* and for an adult female – 2.2 *l/day.* Water performs important functions to support the human body:

- is a vital nutrient for the life of every cell in the human body;
- regulates external body temperature through sweating and breathing;
- metabolizes and transports in the blood carbohydrates and proteins that the human body uses as food;
- physiologically removes waste from the human body through urination;
- lubricates the joints;
- forms saliva.

There are organic, inorganic and microbiological contaminants in the water.

Organic pollution is related to the pollution of water with organic matter. The latter comes from domestic, agricultural or industrial sources. The organic matter is then decomposed by microorganisms and the process is accompanied by the consumption of dissolved oxygen in the water. Toxic metals and organic pollutants have become serious environment issues because these contaminants can cause indelible damage to human health and aquatic organisms [3]. Human activities such as mining, coal burning, and manufacturing (especially battery production) contribute to the discharge of toxic metal pollutants into surrounding water bodies, and there are negative health impacts coming from intake of these pollutants even at low concentrations [4, 5]. Organic pollutants threaten human health and deteriorate water quality due to their biotoxicity and carcinogenesis [6].

Microbiological - the contamination of water with pathogenic bacteria, viruses, some fungi, parasitic worms, etc.

Inorganic pollution is associated with the entry into the water of minerals, chemicals and toxic substances. The most serious sources of inorganic pollutants are the enterprises of the metallurgical, machine-building, ore-mining and coal-mining industries; plants for the production of acids, building materials and mineral fertilizers; the timber industry and the woodworking industry; water transport and others [7].

The composition of natural waters depends on the geo - and biochemical characteristics of the region, on the natural course of physical, chemical and biological processes, as well as on human activity. The chemical composition of natural waters depends on their origin, climatic, chemical, geological and biological characteristics of the region and ongoing processes. Depending on the concentration, the chemical elements are divided into:

Main (> 1 g/l) - Na, K, Mg, Ca,
Cl, S, C, O;

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P, Si, Sr, Mn;

Remains (< 10^{-4} g/l) - Pb, Cd, Ni, Co, Cr, etc. [8].

In recent years, due to the ever-increasing pollution of water from human activity, the need to find a way to its purification and protection is constantly growing. However, some scientists suggest that a "water crisis" is approaching. In order to slow-down and even eliminate the possibility of a water crisis, the following methods of water purification have been developed:

- Physical purification (mechanical, primary);
- Physico-chemical purification;
- Dry cleaning;
- Biological (secondary) treatment;
- Purification (tertiary);
- Decontamination;
- Sludge treatment.

The choice of treatment method depends on the nature of the contamination and the degree of harmfulness of the impurities [9].

When consuming water with indicators above the permissible limits, various pathogenic conditions are observed. With prolonged consumption of water with higher than the acceptable concentrations of arsenic, it accumulates in the thyroid gland and develops endemic goiter. Arsenic also affects the nervous, cardiovascular and respiratory systems, skin and other organs.

Chromium is a carcinogen. According to the World Health Organization Guidelines for Drinking Water Quality (2004), the permissible limit for chromium in drinking water (0.05 mg/l) is indicative only, as there is no conclusive evidence that chromium entering human organism through drinking water causes an immediate health risk [10].

In normal countries for arsenic-contaminated areas drinking water treatment plants are built for the purpose of continuous water quality monitoring and sustainable guarantee of arsenic values in the water below 10 μg/l. For example, to eliminate highly toxic concentrations of arsenic and chromium from water a three-layer filter is constructed, representing ionosorption column [11]. There are also Bulgarian funds for removal of arsenic from polluted waters [12-15].

Nitrates are a major contaminant of groundwater in Bulgaria. Excess levels of nitrates are the most common problem in terms of the physicochemical characteristics of drinking water. Excessive levels of nitrates in the water can cause the development of the disease "water-nitrate methemoglobinemia" in infants and young children. In iodine-deficient areas, it can lead to an increase in the frequency of endemic goiter in adolescents. The methods for reducing nitrates in the waters used for drinking and household needs, known in practice, are ion exchange and reverse osmosis [10].

If the acceptable concentration of fluorides is exceeded, the disease dental fluorosis may be developed, in which fluoride accumulates in the teeth, changes their color, and damages their enamel, which makes them brittle and crumbly. Prolonged exposure to high levels of fluoride through drinking water can also lead to the development of skeletal fluorosis, which affects the entire skeletal system. Several methods are known reducing the content of fluorides in drinking water – filtration by reverse osmosis, distillation filtration, activated carbon filter for defluorination of aluminum and others.

Manganese belongs to the group of heavy metals. Studies show that consuming water with manganese exceeding the acceptable concentration of the metal leads to a decrease in children's intellectual abilities. Constant use of such water can provoke the appearance of serious diseases of the skeletal system. Manganese is an extremely toxic element that has a destructive effect on the nervous and blood systems.

Water chlorination is the most common way to disinfect drinking water, using chlorine gas or chlorine-containing compounds that react with water or salts dissolved in it, but it is also very harmful to the body. The use of chlorinated water during pregnancy can lead to birth defects in children - in particular, heart and brain defects. With prolonged consumption of chlorinated water, different types of diseases develop - cancer, endemic goiter, heart disease, anemia, hypertension and others.

Drinking water is fresh water with a high degree of purity. It must be free of heavy metals, pesticide residues, hormones, antibiotics, bacteria, viruses and other toxic impurities. The content of mineral substances should not exceed the permissible limits [16]. Chlorine compounds are used in the central water supply for disinfection of water and pipelines to destroy harmful microorganisms that cause diseases.

Bulgaria is one of the countries in the world richest in mineral waters of natural origin, with unique composition and drinking qualities. According to the National Register of the Ministry of Health (2019) 22 companies bottle natural mineral waters in Bulgaria [17].

In Bulgaria, the quality of water intended for drinking and household purposes is controlled by Ordinances 9 and 12 on the Quality of water for drinking and household purposes [18, 19]. In recent years, due to the ever-increasing water contamination from human activity, the need for its purification is constantly growing.

There are various methods for water purification:

- Physical purification - used for coarse substances separation;
- Physico-chemical purification - used for finely suspended particles in water;
- Chemical purification - used for pH adjustment and removal of some solutes;
- Biological purification - used for conversion of biological substances from wastewater into biomass;
- Filtration - used for removal of suspended, colloidal and dissolved contaminants;
- Disinfection - used for pathogenic bacteria removal [9].

The hardness of water is due to the calcium and magnesium ions contained in it, usually bound as carbonates. Water hardness (Table 1) is most often expressed as the equivalent of milligrams of $CaCO_3$ per liter or measured in degrees (˚dH). 1˚dH = 17.8 mg / l $CaCO_3$. Hardness can also be determined by what salts remain in the water after boiling. The salts determining the carbonate hardness fall to the bottom as a precipitate. For example, for calcium:

$$Ca(HCO_3)_2 \leftrightarrow CaCO_3\downarrow + H_2O + CO_2\uparrow$$

CO_2 evaporates at boiling, and sparingly soluble $CaCO_3$ precipitates and forms white deposits on the walls of the vessel.

Table 1. Scale for assessing water hardness [24]

$Ca^{2+} + Mg^{2+}$ mg/l	˚dH	Hardness
0-17	0-4	Very soft water
17-60	4-5	Soft water
60-120	9-12	Medium hardness
120-180	19-25	Moderate hardness
>180	>25	Hard water

Sixty years ago, Kobayashi [20] reported a link between water chemistry and the risk of cardiovascular disease [21]. A lot of the data show that harder water has a lower risk of disease and an association may explain why heart disease deaths are more common in the coastal areas of the United States than in the Midwest [22].

Distilled water or demineralized water is that in which the water has been subjected to a treatment which removes all its minerals and salts by reverse osmosis and distillation. This is an absolutely pure form of water, but is not usually recommended for drinking. It can cause mineral deficiency because it is devoid of any salts and most of the natural minerals in the water have been removed as a result of this process. Drinking this water can lead to rapid loss of sodium, potassium, chloride and magnesium [23].

In different regions of Bulgaria, the water has different hardness (see Fig. 1).

The purpose of this research is quality improvement of tap and spring water in Blagoevgrad region for drinking purposes through the use of local purification systems with filtration, reverse osmosis and UV disinfection. Physico-chemical characteristics of water from the water supply network, drilling and spring waters in the Blagoevgrad region were determined. A 7 - stage system with reverse osmosis and UV disinfection for tap water purification was used. Water filtration system and UV disinfection for spring water purification was used too. A cascade system for spring water purification for drinking and technical purposes was constructed and applied.

Reverse osmosis is increasingly used in drinking water supply for treatment of fresh water sources which can directly result in high-quality water. In practice reverse osmosis is never applied directly on fresh water sources, predominantly because of the occurrence of membrane fouling [25].

MATERIALS AND METHODS

Eleven different water types were analyzed (see Table 2). The tap water from Blagoevgrad was additionally purified through a 7-stage reverse osmosis system with UV disinfection (module 1). Some of the waters have been treated by a filtration and UV disinfection system (module 2). Some of the samples also passed through a cascade system (modules 1 and 2).

The groundwater from the villages of Bistritsa, Kovachevica, Blagoevgrad District was not subjected to the systems for additional purification and quality of the driver, as it has very good indicators and no improvement is necessary.

Drilling water from the village of Buchino - 6 m depth was not further purified through the system as it has a very high hardness and electrical conductivity and after additional purification the filters will have to be replaced with new ones because they will be quite dirty.

For the same reason, tap water and drilling water - 55 m depth from the village of Bozduganovo, Stara Zagora District, were not allowed through the systems.

For the bottled Pirinska mineral water "PREDELA" only analyses of the physico-chemical characteristics were made and then they were compared with those of the groundwater from the

"Predela" area. Some of the main physico-chemical characteristics according to Ordinances 9 and 12 for Water quality for drinking and household purposes of these waters were determined: physico-chemical characteristics: total hardness (°dH), Ca^{2+} (mg/l), Mg^{2+} (mg/l), pH; electrical conductivity.

The electrical conductivity was determined with a Hanna DIST 4 EC Tester conductometer with 0.01 µS / cm resolution - HI98303 calibrated with NaCl solution.

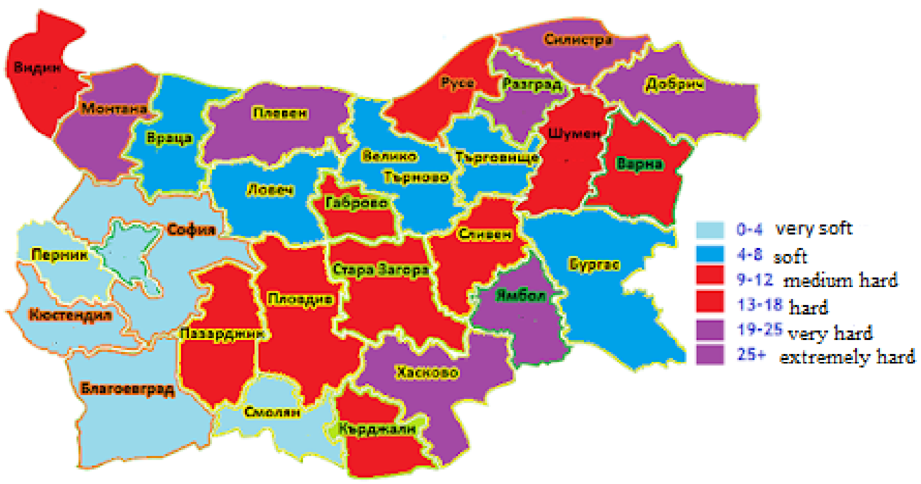

Fig. 1. Water hardness map of Bulgaria, measured in °dH [24]

Table 2. Analyzed waters and systems for improving their quality.

№	Samples	Module 1	Module 2	Module 1 and Module 2
1	Tap water located in Blagoevgrad	yes	-	-
2	Groundwater located in Park „Bachinovo", Blagoevgrad	-	yes	yes
3	Groundwater located in the village of Bistritsa, Kovachevitsa area, Blagoevgrad District	-	-	-
4	Groundwater located in the village of Belo Pole, Blagoevgrad District	-	yes	yes
5	Drilling water located in "Strumsko" Housing Estate, Blagoevgrad, drilling - 11 m	-	yes	yes
6	Drilling water located in the village of Buchino, drilling - 33 m	-	yes	yes
7	Drilling water located in the village of Buchino, drilling - 6 m	-	-	-
8	Tap water located in the village of Bozduganovo, Stara Zagora District	-	-	-
9	Drilling water located in the town of Radnevo, Stara Zagora District, drilling - 55 m.	-	-	-
10	Groundwater (Rilska water) located in the "Predela" area		yes	yes
11	Bottled Pirinska mineral water "PREDELA"	-	-	-

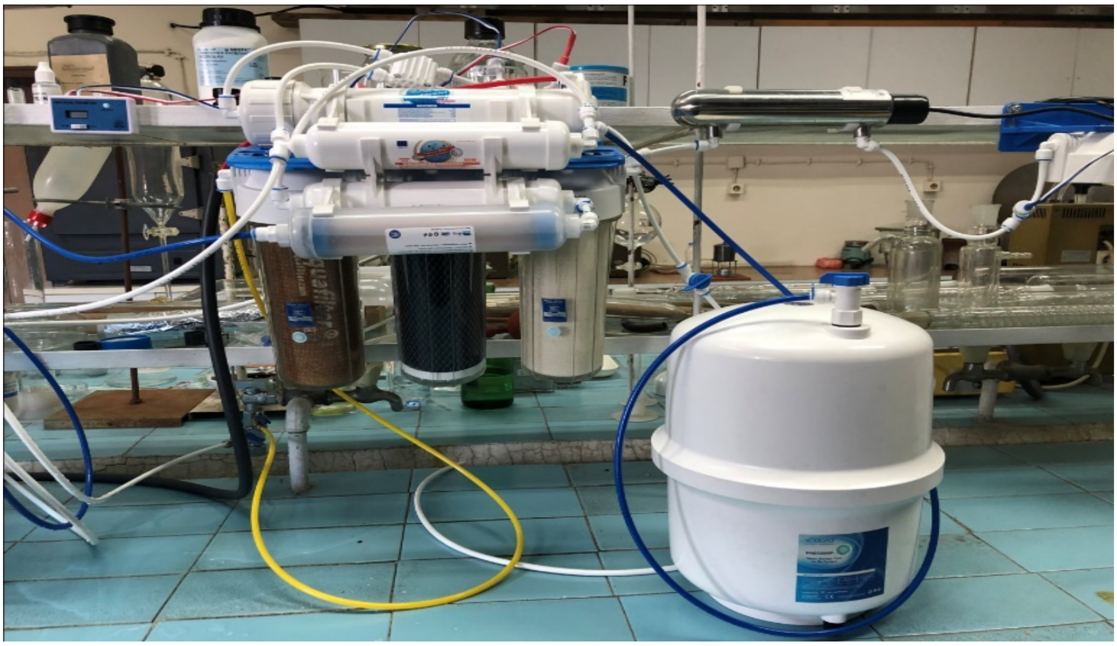

Fig. 2. 7 - stage reverse osmosis system with UV – disinfection (Module 1).

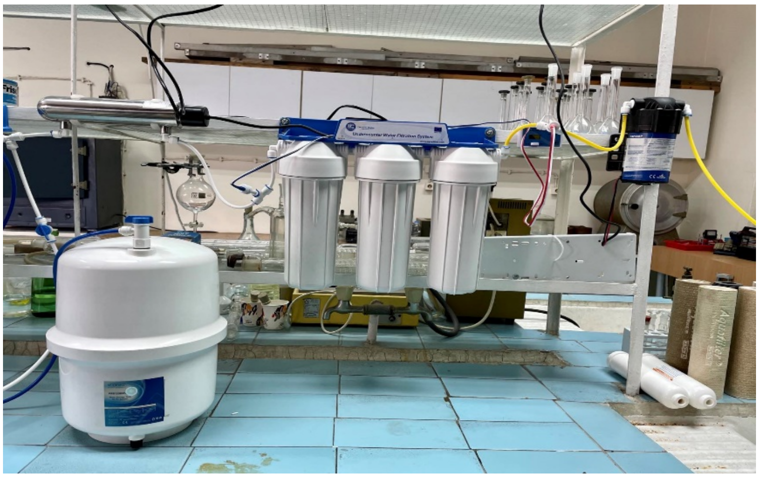

Fig. 3. Spring water purification system with filtration and UV – disinfection (Module 2).

The water hardness is related to the determination of the concentration of Ca^{2+} and Mg^{2+}. First, the total concentration of the two ions was determined. Mg^{2+} was then precipitated in a separate sample by raising the pH and only Ca^{2+} was titrated in this solution. The difference in the two titrations gives the amount of Mg^{2+}. The hardness of water is calculated in German degrees by the formula:

$$\frac{V_T\ N_T EM_{CaO}}{V_{\mathrm{pr}}}.100$$

where V_T is the volume of the titrant; N_T is the normality of the titrant; EM $_{CaO}$ - the equivalent mass of CaO; Vpr - the volume of the sample. pH was measured on a Hanna pH meter. Reverse osmosis system with UV disinfection was used (see Fig. 2). Additional information is presented in Supplementary.

This sytem was delivered to the Department of Chemistry, part of Faculty of Mathematics and Natural Sciences at the South-West University "Neofit Rilski" by "Pavirani" company. The 7-stage reverse osmosis system with UV disinfection purifies the water from all available contaminants [26].

With the three pre-filters - a 20 micrometer filter, an activated carbon filter and a 5 micrometer filter, the water is purified from sludge and chlorine and prepared for fine filtration with a reverse osmosis membrane. After passing through the membrane, the water is clean, without chemical compounds, without heavy metals and other substances. Up to this stage the so-called technical water is obtained - it has not passed through the mineralizer. It is suitable for technical purposes. In the next stage, the water passes through a filter of activated carbon from coconut shells, which aims to remove chlorine, pesticides, and organic compounds. The mineralizer enriches the water with useful minerals such as calcium and magnesium, giving the water the qualities of slightly mineralized table water. The system also contains an ionizer, a UV sterilizer and a TDS meter [26, 27]. A spring groundwater purification system with filtration and UV disinfection, which consists of a reverse osmosis diaphragm pump, necessary to supply water to the system through a certain pressure; filter column with mechanical filter of 20 and 5 micrometers, activated carbon filter, UV lamp, TDS meter and tank for collecting purified water are presented in Fig. 3. Characteristics of the diaphragm pump for reverse osmosis and of grid tie PV generator are presented in Supplementary.

Figure 4 shows the cascade system (consisting of two modules – Module 1 and Module 2) for spring water purification, which is assembled by us. It is a combination of the two systems - a system with filtration and UV disinfection and a purification system with reverse osmosis. The electricity consumed for the pump and UV lamps is "green" from building integrated photovoltaic systems to the laboratory. Thus, such a water purification system together with the photovoltaic system can be used in regions that are isolated from the energy system.

Fig. 4. Cascade spring water purification system

RESULTS AND DISCUSSION

The results of Blagoevgrad tap water (without prior purification), water purified with a 7-stage reverse osmosis system for obtaining drinking and technical water and tap water from Bozduganovo are presented in Fig. 5. The physico-chemical indicators - total hardness (˚dH), Ca^{2+} (mg/l), Mg^{2+} (mg/l), pH and the electrical conductivity are compared.

The technical water is a better solvent for detergents, better extracts coffee and tea and leaves no traces after washing glass surfaces.

From the analyses made technical water has the lowest hardness (lowest content of Ca^{2+} and Mg^{2+}) and conductivity compared to the other two waters. This result is related to the fact that the technical water hasn´t passed through the mineralizer, but only through the three filters and the reverse osmosis membrane.

The water from the pipeline in Blagoevgrad is soft. After passing through the system, it is enriched with Ca^{2+} and Mg^{2+} and purified.

The wastewater during the treatment of water from the pipeline in Blagoevgrad has low mineralization and this allows it to be collected and fed into the cascade system for additional treatment. This saves water.

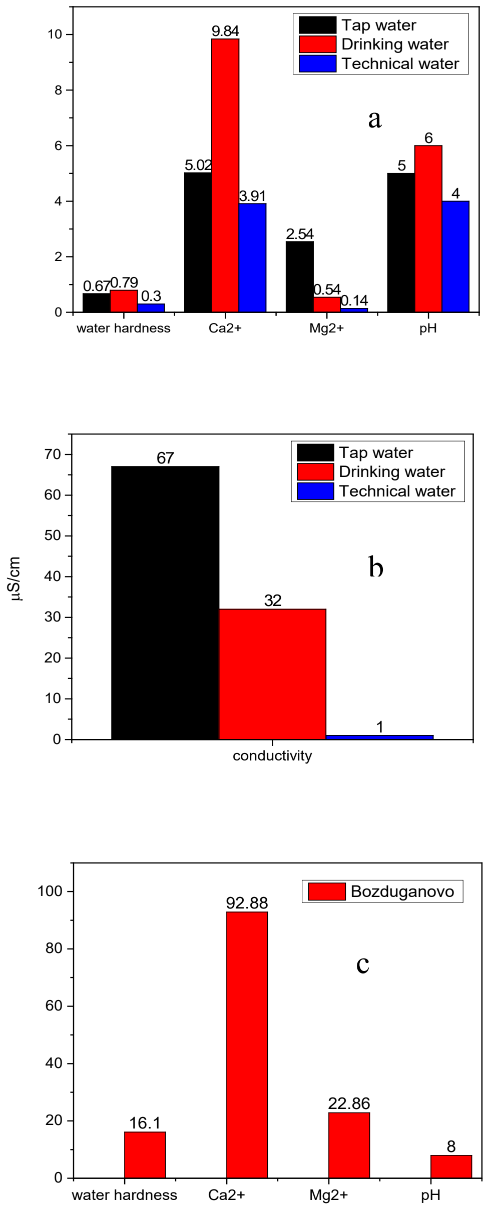

Fig. 5. Blagoevgrad tap water (without prior purification), purified with a 7-stage reverse osmosis system for obtaining drinking and technical water: a) Comparison of physico-chemical characteristics; b) Comparison of the electrical conductivity; c) Tap water from Bozduganovo – physico-chemical characteristics.

There is a significant difference in the content of Ca^{2+}, Mg^{2+}, total hardness and pH of tap water from Blagoevgrad and Bozduganovo. The high levels of these physico-chemical characteristics of the tap water in the village of Bozduganovo in comparison with those of the tap water from Blagoevgrad show that the water is much harder.

The results of the physico-chemical characteristics and the electrical conductivity research of groundwater located in the village of Bistritsa, from Park "Bachinovo" and the village of Belo Pole are presented in Figure 6. The groundwater from the village of Bistritsa has the best characteristics. It is softer compared to the water from "Bachinovo" and the village of Belo Pole due to the lower levels of Ca^{2+} and Mg^{2+}. The water from the village of Bistritsa has the lowest electrical conductivity again, which makes it best for drinking and for technical purposes in comparison with the other two water sources.

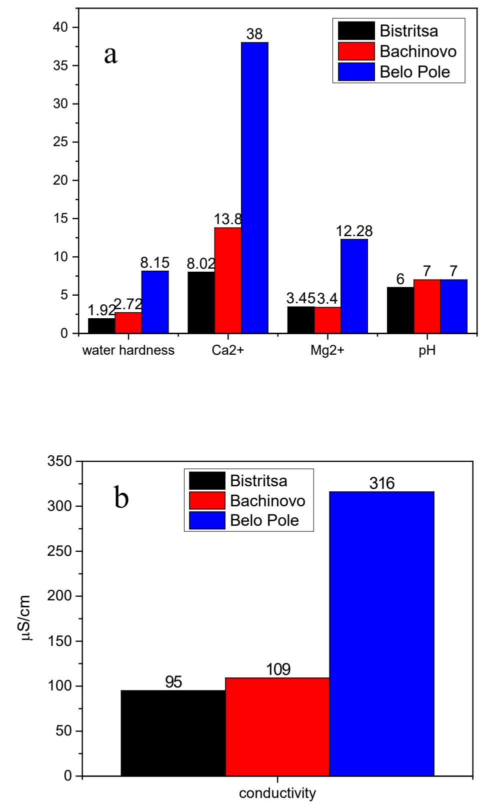

Fig. 6. Groundwater water*:* a) Comparison of physico-chemical characteristics; b) Comparison of the electrical conductivity.

The analyses of groundwater drilling water located in the village of Buchino - 33 m depth of the incoming water, revealed that the water has a high electrical conductivity, a higher content of magnesium than calcium ions and a high mineralization – medium-hard water. This water is suitable for drinking needs for daily consumption. Figure 7 shows the results of the physico-chemical characteristics - total hardness (˚dH), Ca^{2+} (mg/l), Mg^{2+} (mg/l), pH analyses of drilling waters taken from different sources. The water from groundwater drilling in the village of Buchino - 6 m depth is the hardest one and the water taken from the groundwater drilling in the town of Radnevo (55 m depth) is the softest due to several factors. For instance: deep water is cold and the solubility of salts is lower than in other water, which is at a higher level. Water up to 6 m is more suitable for watering than for drinking purposes, as there may be various impurities due to soil fertilization. The EU is considering a ban on the use of manure, as there are no guaranteed ingredients, i.e. it may contain heavy metals, which can lead to water contamination. And to impose the use of fertilizer, which has a certain composition.

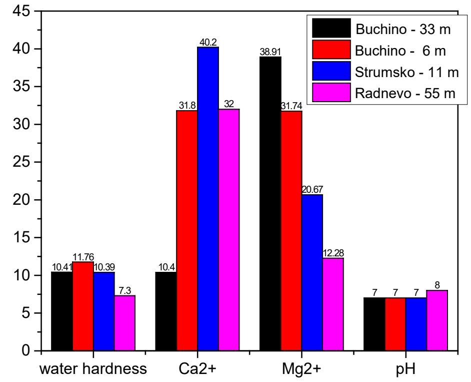

Fig. 7. Drilling groundwater - physico-chemical characteristics of a water located in the village of Buchino, "Strumsko" Housing Estate, Town of Radnevo.

The groundwater water in Predela area is softer compared to the Predel mineral water due to the higher levels of Ca^{2+} and Mg^{2+} (see Fig. 8). The electrical conductivity depends on the water mineralization; the water purity is judged by the electrical conductivity. For this purpose, there are conductometers mounted on the system.

The conductivity of different water types (tap water located in Blagoevgrad, tap water located in the village of Bozduganovo, drilling located in the village of Buchino - 33 m depth, drilling located in the village of Buchino - 6 m depth, drilling located in "Strumsko" Housing Estate - 11 m, drilling located in the town of Radnevo - 55 m depth, groundwater from "Predela" area and bottled mineral water "PREDELA") are presented in Figure 9.

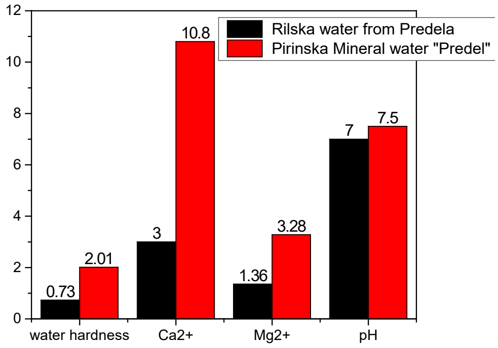

Fig. 8. Comparison of physico-chemical characteristics of groundwater.

The drilling water from the town of Radnevo (55 m depth) has the highest electrical conductivity, and the Pirin groundwater water from the "Predela" area has the lowest one.

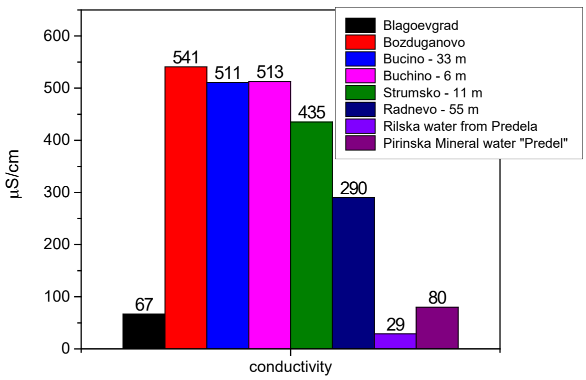

Fig. 9. Comparison of the electrical conductivity.

CONCLUSION

Local purification of tap water with a 7 - stage reverse osmosis system with UV disinfection improves its quality for drinking and technical purposes. The quality of groundwater for drinking purposes is improved by means of a local purification system with filtration and UV disinfection. By using a cascade system with a pump, powered by a photovoltaic generator in isolated areas of the energy system, local purification of groundwater for drinking purposes with initial and lower mineralization, as well as technical water is achieved. The most suitable waters for drinking purposes in Blagoevgrad Region are:

(i) Groundwater located in the villages of Bistritsa, Kovachevitsa, Blagoevgrad District;
(ii) Groundwater located in Park „Bachinovo", Blagoevgrad;
(iii) Groundwater (Rilska water) located in the "Predela" area;
(iv) Bottled Pirinska mineral water "PREDELA";
(v) Blagoevgrad tap water after local purification at home.

Suitable for drinking purposes, from drilled ground waters, after local filtration and UV disinfection are those located in the town of Radnevo (55 m depth) and in the village of Belo Pole (33 m depth).

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