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The analysis of chemical parameters of groundwater before and after sand filtration in the Velekinca water treatment plant, Kosovo

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Abstract

Our scientific research is based on the monitoring of ions before and after filtration of groundwaters in the water plant of Velekinca in the municipality of Gjilan, Kosovo. Sandy filters are the most commonly used industrial filters in surface – and groundwater industries. The reason is their low construction cost and high processing capacity. In our scientific research, sand filters used in the plant do not have perfect filtration, so we can monitor results before filtration (BF) and after filtration (AF) by determining the concentration of some ions and molecules. The following average concentrations have described: Ca^{2+} (BF: 83.42, AF: 83.19) mg·dm⁻³, Mg²⁺ (BF: 35.59, AF: 34.35) mg·dm⁻³, Cl⁻ (BF: 28.018, AF: 28.73) mg·dm⁻³, SO₄²⁻ (BF: 42.76, AF: 44.46) mg·dm⁻³, HCO₃⁻ (BF: 410.9, AF: 404.81) mg·dm⁻³, A-HCl (BF: 6.73, AF: 6.63) ml-HCl, GH (BF: 19.94, AF: 19.62) °dH, CS (BF: 18.87, AF: 18.5) °dH and NO₂⁻ (BF: 0.0033, AF: 0.0022) mg·dm⁻³. Being scientific researchers in the field of water treatment technology, we have concluded that ions create an affinity for sand particles. They attach to each other by creating an ion-sand particle physical chain. According to our scientific research, sand filters are difficult to guarantee a high quality of water processing.

Key words: groundwater treatment plant, ion-sand particle chain, physical-chemical parameters, sand filtration, WHO standard

INTRODUCTION

Water diversion is a globally historic and popular engineering method for water supply [DAI *et al.* 2018]. Water security is of paramount concern, particularly in countries facing significant population growth and a drying climate [BEKELE *et al.* 2018]. The analysis of water produced is critical to monitor field operation, control processes, evaluate appropriate management practices and treatment effectiveness, and assess potential risks to public health and environment during the use of treated water [JIANG *et al.* 2021]. Groundwater chemistry provides enormous vital information on suitability for domestic, irrigational and industrial purposes, and it is very helpful in understanding and identifying processes determining the hydrochemical quality of groundwater [ALAM 2014; XIAO *et al.* 2017]. Rare earth elements concentrations in groundwater vary depending on specific water-rock interactions and the presence of dissolved or colloidal organic and inorganic species, etc. [DIA *et al.* 2000; LIU *et al.* 2018]. Since high concentrations of ions signal certain problems, water to be clean should be physically and chemically processed.

When transferred from laboratories to industrial applications, processes need to be more durable in operation and production. This has been accomplished by a growing number of operators in the industry who carefully followed and collected process data. The cost of the process, ever larger capacity of plants, and difficult working conditions have forced the creation of a new engineering discipline, which today is called a galopant development. In order to generate a normal performance of industrial processes, a set of measurements and adjustments need to be performed on working chemical or physical parameters [PIN-GULI *et al.* 2017]. Our research is based in the field of environmental science, i.e. quality of groundwater in the groundwater treatment plant (GWTP). Sand filters have been our main focus because they accumulate different chemical ions that can cause problems in water processed for direct consumption.

Both calcium and magnesium are essential to human health. Inadequate intake of either nutrient can impair health [COTRUVO, BARTRAM (eds.) 2009]. The presence of the soluble Ca^{2+} and Mg^{2+} salts causes unsuitable behaviour of hard water solutions for drinking, watering, and industrial purposes [CETIN 2014; VIERO *et al.* 2002]. Hard water is usually defined as water which contains a high concentration of calcium and magnesium ions. Carbonate hardness is sometimes called temporary hardness because it can be removed by boiling. Non-carbonate hardness (carbon strength) cannot be broken down by boiling of water, so it is also known as permanent hardness etc. [BE-LULI 2017].

Chlorides are the most widespread anions in surface and groundwater, and the concentration of chlorides in natural waters varies. Chlorides are not considered very desirable, but in most cases its concentration from 70-150 mg·dm⁻³ causes serious problems for humans and plants [DACI, DACI-AJVAZI 2014]. Sulphate ions, as well as chloride ions, are found in all-natural waters. In groundwater, SO₄ content is generally higher than in rivers and lakes. The main source of sulphate ions in water is gypsum [SHEHU 2009]. The main source of nitrite ions (NO₂-N) in water is the process of mineralization of organic matter and nitrification from bacteria [BELULI 2019]. Nitrite is the ionic intermediate state between nitrate and ammonia nitrogen, which explains their low quantities encountered in the aquatic environment [KADAOUI et al. 2019]. However, studies on acute and chronic effects of nitrite in various parts of the world have shown that their different concentrations in water and nutrients lead to several diseases in different population groups [PARVIZISHAD et al. 2017]. Groundwater can traverse the surface layers of soil until it reaches water accumulation areas, which are indescribable rock layers. Often, these are limestone formations but also gypsum, clayey, etc. Water that has been in contact with limestone layers is enriched with HCO₃⁻ [BELULI et al. 2017].

After that, we have analysed chemical parameters such as calcium (Ca²⁺), magnesia (Mg²⁺), chlorides (Cl⁻), general hardness (GH:°dH), carbon strength (CS:°dH), alkaline (A:HCl), bicarbonate (HCO₃⁻), and nitrite (NO₂-N).

Table 1. Permissibility criteria for physical-chemical parameters according to WHO

Parameter	Symbol	Measurement unit	WHO standard
Calcium	Ca ²⁺	mg∙dm ⁻³	<200
Magnesium	Mg ²⁺	mg∙dm ⁻³	<50
Chlorides	Cl	mg∙dm ⁻³	<200
Nitrites	NO ₂ -N	mg∙dm ⁻³	0.6
Bicarbonates	HCO ₃ ⁻	mg∙dm ⁻³	630
Alkaline	A-HCl	cm ³	10.5
General hardness	GH:°dH	-	30
Sulphate	SO_4^{-2}	mg∙dm ⁻³	<200

Source: WHO [1972].

These chemical parameters have been analysed in the groundwater analytic laboratory and are compared with data provided by the World Health Organization (WHO), see Table 1.

STUDY METHODS

SAMPLING IN THE GROUNDWATER TREATMENT PLANT (GWTP)

Sampling techniques are relatively important in analytic chemistry, especially when samples do not have so much suspended solids (e.g. drinking water samples). The volume of samples is good to be 0.5–2.0 dm³ [ÇULLAJ 2010]. Water samples in our scientific research have been determined in the laboratory of analytical and instrumental chemistry, so the samples are not transported outside the industry because all scientific research is based inside the industrial facility and the discussion of results and experimental part is discussed and monitored by experts for water technology (Figs. 1, 2).







Fig. 2. Analysis of samples before and after filtration in groundwater treatment plant; source: own elaboration

SAND BED FILTER DESIGN IN THE WATER TREATMENT PLANT IN VELEKINCA

Regardless of the application, the recommended depth of the sand is around 0.6–1.8 m [COULSON *et al.* 1991]. The sand bed depth in the two water treatment plants is 1.5 m.

Dimensions of the sand filters are as follows: width 2 m and length 4 m (Photo 1). A compromise is that most rapid pressure sand bed filters use sand grains of 0.6-1.2 mm.

Photo 1. Sand filters in the water treatment plant in Velekinca (phot. V. Beluli)

The effective diameter (D_E) and the uniformity coefficient (C_U) are two important granulometric characteristics of a filter material. The coefficient of uniformity is the ratio between the diameter that allows the passage of 60% of particles and the one that allows the passage of 10%; the proportion D_{60} : D_{10} . The main elements of these filters include the end of the filter, gravel support and the filter medium. The end of the filter is a structure that separates the filter medium from filtered water. It is stable and it holds the filter medium (1 m sand and gravel) and the water that is above the filter medium. It also allows to collect and remove filter water and uniform distribution of washing water [OSMANAJ, LAKO 2017], see Figure 3.

The gravel support is place on the bottom of the filter. It keeps the sand of the filter medium and regulates the distribution of washing water in the filter. The filter medium contains very fine sand of 0.4–1.0 mm in diameter.

CHEMICALS

Ethylenediaminetetraacetic acid (EDTA), Nitri Ver 3 reagent (HACH®), Sulfa Ver 4 (HACH®), AgNO₃ ($C = 0.01 \text{ mol} \cdot \text{dm}^{-3}$), HCl ($C = 0.01 \text{ mol} \cdot \text{dm}^{-3}$), buffer, indicator (black erythromycin), indicator (methylorange), indicator (phenolphthalein), indicator (black murexide), NaOH ($C = 2 \text{ mol} \cdot \text{dm}^{-3}$), methylene chloride, K₂Cr₂O₇, H₂SO₄ 98%, glucose.



Fig. 3. Detailed structure of the sand filter in the water treatment plant in Velekinca: *I* = water, *2* = fine sand, *3* = thick sand, *4* = filter support, *5* = filter end, *6* = filter body; source: HIDROMORAVA [2019]

CHEMICAL PARAMETERS AND METHODS OF ANALYSIS

Absorption spectrometry is based on the electromagnetic radiation absorption by molecules in the UV spectra of 160-400 nm (ultraviolet) and VIS 400-780 nm (visible). UV-VIS radiation absorption causes the excitation of electrons in chemical bonds by pushing molecules to higher energy levels [VASJARI et al. 2013]. The absorption of UV--VIS radiation from complex molecules and inorganic salts of transitional metals, as well as of lanthanides and actinides, causes the molecule to move from its basal to its excited state [BELULI 2018]. The HACH® Model DR/2010 Spectrophotometer is a microprocessor-controlled singlebeam instrument for colorimetric testing in the laboratory or in the field [Hach 1999]. The instrument is precalibrated for over 120 different colorimetric measurements and allows convenient calibrations for user-entered and future HACH methods.

Sulphate (SO₄²⁻) concentration has been determined using Sulfa Ver 4 (0–70 mg·dm⁻³), method 8051), and the absorbance level is then measured using a spectrophotometer (HACH[®] DR/4000) at $\lambda = 450$ nm. Nitrite (NO₂-N) concentration is determined using Nitri Ver 3 reagent (test 0–0.300 mg·dm⁻³, method 8507), and the absorbance level measured using a spectrophotometer (HACH[®] DR/2010) at $\lambda = 507$ nm.

Water hardness or general hardness (*GH*) is analysed by adding 2–5 cm³ of buffer and indicator (black erythromycin) in very small quantities to a sample of 100 cm³ of water. Following the addition of the indicator, the solution becomes red or light red, and the titration is done with complexon III or EDTA ($C_{EDTA} = 0.01 \text{ mol} \cdot \text{dm}^{-3}$) until the solution changes its colour to intensive blue [BELULI 2018]. The calculation is based on Equation (1):

$$GH(^{\circ}dH) = V_{EDTA} \cdot C_{EDTA} \cdot 56 \cdot 1000 / V_s$$
(1)

Where: V_{EDTA} = the titration volume (cm³) with ethylenediaminetetraacetic acid, C_{EDTA} = the concentration of EDTA ($C_{\text{EDTA}} = 0.01 \text{ mol} \cdot \text{dm}^{-3}$), V_s = the volume of the sample used.

Carbonate strength (*CS*) is defined as the alkalinity to methylorange. A volume of 100 cm³ water sample was transferred to 500 cm³ Erlenmeyer flask and 2–3 drops of methylene chloride were added. The titration was performed with standard solution HCl ($C = 0.01 \text{ mol} \cdot \text{dm}^{-3}$) until the colour changed to orange [BELULI 2018; 2019]. The analysis results were calculated in German degrees (°dH) water hardness scale according to Equation (2)

$$CS(^{\circ}dH) = 2.8 V_{HCl} \cdot C_{HCl}$$
(2)

Where: 2.8 = the constant, V_{HCl} = consumed volume of HCl, C_{HCl} = the concentration of HCl ($C = 0.01 \text{ mol} \cdot \text{dm}^{-3}$).

First, we get 100 cm³ of water, add 5 cm³ of buffer solution – in this case NaOH ($C = 2 \text{ mol} \cdot \text{dm}^{-3}$) and a black murexide indicator (black murexide is prepared from ammonia purities mixed with NaCl ($C = 0.01 \text{ mol} \cdot \text{dm}^{-3}$), and titrated with EDTA ($C = 0.01 \text{ mol} \cdot \text{dm}^{-3}$) to change the colour from red to purple. The titration should be carried out for 5 min after the addition of NaOH [BELULI 2018]. The determination of Ca²⁺ (in mg·dm⁻³) is calculated by Equation (3):

$$Ca^{2+} = V_{EDTA} \cdot C_{EDTA} \cdot 56\ 1000 / V_s \tag{3}$$

Where: V_{EDTA} = the volume (cm³) of the titre with ethylenedi-aminetetraacetic acid, C_{EDTA} = the concentration of EDTA ($C = 0.01 \text{ mol} \cdot \text{dm}^{-3}$), V_s = the volume of the sample used.

The content of Mg^{2+} (mg·dm⁻³) is calculated with Equation (4) [BELULI 2018]:

$$Mg^{2+} = \frac{GH/^{\circ}dH - Ca^{2+}}{10} 7.19$$
 (4)

Water sample alkalinity (A) is the measurement of its capacity to neutralize acids. Four drops of phenolphthalein are added to 100 cm³ of the sample. If the 100 cm³ solution becomes purple, the pH of water contained bases is above 8.3, and if the solution does not turn purple, 2–3 drops of methylorange are added, which turns the solution yellow. The solution is then titrated with HCl (C = 0.01 mol·dm⁻³) until it turns orange, and the amount of titre used is recorded.

The determination of chlorides was carried out in an Erlenmeyer flask containing 100 cm³ of water sample (adjust pH 7–10 if necessary). With the addition of 1 cm³ of K₂CrO₄ 0.257 mol·dm⁻³, the sample turned yellowish. Titration was done with silver nitrate (AgNO₃ (C = 0.01 mol·dm⁻³)) and it stopped when the solution got light red [BELULI 2018; 2019]. The value of chlorides (mg·dm⁻³) in the sample was calculated according to Equation (5):

$$Cl^{-} = 35.453 c (V_1 - V_2) / V_s$$
(5)

Where: V_1 = the volume of the titre for the sample (cm³), V_2 = the volume of the titre for blind sample (cm³), c = molarity AgNO₃ (C = 0.01 mol·dm⁻³), V_s = the volume of the sample used (100 cm³ in our research).

RESULTS AND DISCUSSION

Results for calcium (Ca^{2+}). The Ca^{2+} ion in groundwater varies from 76.55 to 90.58 mg·dm⁻³ (Fig. 4. The problem exists in water filtration where Ca^{2+} concentration increases after water filtration. The concentration of Ca^{2+} before and after the filtration of groundwater has changed and we have divided them into two groups based on their concentrations:

- a) in samples SP₁, SP₃, SP₈, SP₁₀, SP₁₃, SP₁₄, SP₁₅ and SP₂₀, the concentration of Ca²⁺ in water after filtration is more vivid and varies from 0.98 to 7.2 mg·dm⁻³ (Fig. 4);
- b) in the samples SP₂, SP₄, SP₅, SP₆, SP₇, SP₉, SP₁₁, SP₁₂, SP₁₆, SP₁₇, SP₁₈ and SP₁₉, Ca²⁺ concentration after filtration varies from 0.8 to $3.61 \text{ mg} \cdot \text{dm}^{-3}$ (Fig. 4).

Lab results show that in the first group of calcium samples it has not been reduced after filtration but the concentration of calcium is more present. Increasing the concentration of Ca^{2+} in water after the filter does not pose any risk because the concentration is still in accordance with the WHO regulation (Tab. 1). In the second group of samples, the concentration of Ca^{2+} is reduced but not much.

Results for magnesium (Mg^{2+}). In our research, the magnesium concentration (Mg^{2+}) as $mg \cdot dm^{-3}$ is not very high and is in line with the WHO (Tab. 1, Fig. 4). The concentration of Mg^{2+} in the GWPT after analysing a large number of water samples before filtration is from 30.5 to 41.7 mg \cdot dm^{-3} (Fig. 4). The concentration of Mg^{2+} after filtration is from 28.3 to 40.1 mg \cdot dm^{-3}, specifically in our research in the GWPT after water filtration, different concentrations of Mg^{2+} result from changes after filtering and we have divided them into two groups:

- a) in SP₃, SP₅, SP₈, SP₉, SP₁₁, SP₁₄, SP₁₆, SP₁₈ and SP₂₀, the concentrations of Mg²⁺ has not been reduced, so the concentration of Mg²⁺ increased after filtration from 1.08 to 4.32 mg·dm⁻³ (Fig. 4);
- b) in SP₁, SP₂, SP₄, SP₆, SP₇, SP₁₀, SP₁₂, SP₁₃, SP₁₅, SP₁₇ and SP₁₉, the concentration of Mg^{2+} is reduced from 0.04 to 3.4 mg·dm⁻³ (Fig. 4).

Results for alkalinity (A-HCl). Alkalinity refers to the capability of water to neutralize acid. A-HCl as a chemical parameter in this research displays no risk and at all times the alkaline concentration is from 6.1 to 7.2 cm^3 and it is in compliance with the WHO (Fig. 4, Tab. 1). A-HCl concentrations before and after the filtration of groundwater has changed and we have divided them into two groups:

- a) in samples SP₅, SP₆, SP₉, SP₁₀ and SP₁₁, the concentration of alkaline after filtration varies with increasing concentration from 0.1 to 0.3 cm³ (Fig. 4);
- b) in the SP₁, SP₂, SP₃, SP₄, SP₇ and SP₈ samples, the concentration in filtered water varies with a decrease in concentration from 0.1 to 0.7 cm³ (Fig. 4).

Results for general hardness (*GH*)**.** In the GWPT, *GH* is in accordance with the WHO and it is classified as hard water of 18.53 to 21.67 °dH. Values of *GH* before and after filtration have changed and we have divided them into two groups:

- a) in SP₇ and SP₈ samples, *GH* after filtration varies 0.74 to 0.95 °dH (Fig. 4); these values are not very high if we compere to other chemical parameters analysed and they exhibit higher levels after filtration;
- b) in the samples SP₁, SP₂, SP₃, SP₄, SP₅, SP₆, SP₉, SP₁₀ and SP₁₁, *GH* after filtration decreases and varies from 0.39 to 0.73 °dH (Fig. 4).















Fig. 4. Results of chemical parameters before and after water filtration in groundwater treatment plant: calcium (Ca²⁺), magnesium (Mg²⁺), chlorides (Cl⁻), sulphates (SO₄²⁻), bicarbonates (HCO₃⁻), alkalinity (A-HCl), general hardness (*GH* – °dH), carbonic strength (*CS* – °dH) and nitrite (NO₂⁻); source: own study

17.0



Fig. 5. Connection of ions in water during filtration from sand filters; source: own study

Results for carbon strength (*CS*). The carbon strength (*CS*/ $^{\circ}$ dH) means a permanent hardness of water. In our research, *CS* is from 17.1 to 20.4 $^{\circ}$ dH (Fig. 4). *CS* values before and after the filtration of groundwater have changed and we have divided them into two groups:

- a) in the SP₅, SP₆, SP₁₀ and SP₁₁ samples, CS values after filtration increase and vary from 0.3 to 0.6 °dH (Fig. 4);
- b) in the SP₁, SP₂, SP₃, SP₄, SP₇, SP₈ and SP₉ samples, CS values after filtration decrease and varies from 0 to 0.73 °dH (Fig. 4).

Results for bicarbonates (HCO₃⁻). In our scientific research, the concentration of HCO_3^- is from 372 to 439 mg·dm⁻³ (Fig. 4). The concentration of HCO_3^- before and after the filtration of groundwater has changed and we have divided its values them into two groups:

- a) in samples SP₅, SP₆, SP₉, SP₁₀ and SP₁₁, the HCO₃⁻ concentration increases and varies from 6.1 to 12 mg·dm⁻³ (Fig. 4);
- b) in samples SP₁, SP₂, SP₃, SP₄, SP₇ and SP₈, the concentration of HCO_3^- decreases and varies from 6.1 to 42 mg·dm⁻³ (Fig. 4).

Results for sulphates (SO₄²⁻). Sulphates as a chemical parameter in our scientific research has been determined because sulphates are often present in many groundwaters. The concentration of SO_4^{2-} is from 39.04 to 46.2 mg·dm⁻³ in the groundwater treatment plant (GWPT). Concentration values are in accordance with WHO (Tab. 1, Fig. 4). The samples in the GWPT before and after filtration are divided into two groups:

- a) in samples SP₁, SP₃, SP₄, SP₅, SP₆, SP₇ and SP₉, the concentration of SO₄^{2–} after filtration increases and varies from 0.4 to $5.12 \text{ mg} \cdot \text{dm}^{-3}$ (Fig. 4);
- b) in samples SP₂, SP₈, SP₁₀ and SP₁₁, the concentration of SO_4^{2-} decreases and varies from 0.18 to 3.0 mg·dm⁻³ (Fig. 4).

Results for nitrites (NO₂⁻). Nitrite (NO₂⁻) as a chemical parameter is necessary for analysis in surface and groundwaters. Its high concentration displays major problems as explained above. The amount of NO₂⁻ in our research is from 0.0025 to 0.004 mg·dm⁻³ (Fig. 4). In sam-

ples from the GWPT before and after filtration, the NO₂⁻ concentration is in accordance with the WHO (Tab. 1, Fig. 4). The NO₂⁻ reduction after filtration is from 0 to $0.002 \text{ mg} \cdot \text{dm}^{-3}$. After filtration, the concentration of NO₂⁻ has not increased in comparison to some other chemicals.

Results for chlorides (CI⁻). Chlorides before and after filtration are determined to compare the concentration of chloride ion reduction. In our case, the concentration of Cl⁻ is from 25 to $30.01 \text{ mg} \cdot \text{dm}^{-3}$ and it is in accordance with the WHO regulation (Fig. 4, Tab. 1). Cl⁻ concentration values before and after the filtration of groundwater have changed and we have divided them into two groups:

- a) in samples SP₂, SP₆ and SP₈, the concentration of Cl⁻ after filtration increases and varies from 0.4 to 3.0 mg·dm⁻³ (Fig. 4);
- b) in the SP₁, SP₃, SP₄, SP₅, SP₇, SP₉, SP₁₀ and SP₁₁ samples, the concentration of Cl⁻ decreases and varies from 1.0 to 2.5 mg·dm⁻³ (Fig. 4).

Being scientific researchers in the field of water treatment technology, we have concluded that ions create an affinity for sand particles and they attach to each other by creating an ion-sand particles and a physical chain. This is caused by not cleaning the filters often or not cleaning the sand filters well. When water flows into the filter at a certain speed that depends on the daily processing capacity, the water flow captures ions that remain in the filter and increases the concentration of ions after filtration (Fig. 5).

CONCLUSIONS

Our scientific research expresses a problem with sand filters that do not guarantee water filtration quality. According to this study, sand filters cannot guarantee a high quality of water processing, as it was the case in the groundwater treatment plant (GWPT). We have examined the question of which ions are the least reduced during the filtration of groundwater. According to the study, the reduction in sand filters of Ca^{2+} and Mg^{2+} ions was insufficient. Therefore, we conclude that when the water flow is larger ions do not stay long in a filter as opposed to a slow-

er flow of water when the concentration of ions inside the sand filter increases after filtration:

- the concentration of Ca²⁺ after water filtration increased from 1 to 7 mg·dm⁻³;
- the concentration of Mg²⁺ after water filtration increased from 1 to 4 mg·dm⁻³;
- the concentration of HCO₃⁻ after filtration in water increased from 6 to 12 mg·dm⁻³;
- the concentration of SO_4^{2-} after filtration in water increased from 0.4 to 5 mg·dm⁻³;
- the concentration of Cl^- after filtration in water increased from 0.4 to 3 mg·dm⁻³.

The concentration of A-HCl, *GH*, *CS* is not taken into account because its pre- and after filtration differences are too low. The second conclusion refers to another scientific study. Sand filters are cleaned approximately every 200 hours of operation. We suggest sand filters to be cleaned after 150 hours because when the filter works nearly 160 hours, the concentration of ions starts to increase gradually. The results of this analytical study have enabled us to understand that the time interval for cleaning sand filters with air is very important because, as shown by analytical results before and after filtration, there is an insufficient reduction of ions. The study helps us to increase the quality of water treatment but it has also been a scientific factor for many water industries in Kosovo.

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