



PROFESSIONAL PAPER

Causes of increased concentration of fluorides in groundwater in Srebrenica municipality

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Abstract

Groundwater, as the main resource of drinking water in the Republic of Srpska, is sometimes neglected regarding quality monitoring. It is especially characteristic of rural water supply sources as autonomous waterworks, individual intakes and wells. For the first time comprehensive analyses of the groundwater quality in rural areas of Srebrenica municipality have been provided in 2010. The results are more than interesting. Beside some standard problems in physico-chemical properties like increased turbidity, iron and manganese, there was emerged problem of increased fluoride concentration in more than one third samples of groundwater. The study of fluoride concentration, given in this paper, includes the causes of this phenomena and possible health impacts. It defines geological setting, or precisely mineralogical properties of the rocks and their interaction with groundwater, as a key factor of the problem. Further, analyses include recommendations regarding future exploration areas with probably same effect of the geological setting on groundwater quality in the territory of the Republic of Srpska. It is very important because the problems related to the long-term consumption of groundwater with fluoride concentration above 1.2 mg/L could be a trigger for very serious diseases like dental and skeletal fluorosis, diseases of pineal gland, kidneys, urinary bladder, thyroid gland, gastro-intestinal tract etc.

1. INTRODUCTION

Groundwater represents the main source of drinking water worldwide, in the Republic of Srpska as well.

In accordance with the official data only about 48% of the population in the entity is covered by public waterworks. The rest of the population (52%) provides drinking water from autonomous waterworks and individual wells or intakes [1].

The level of groundwater quality control is tremendously different between public and other drinking water sources.

In accordance with the Law on Water [2] and the Rule on Health Correctness of Drinking Water Quality [3] (further: the Rule) there is just obvious duty of drinking water quality control of public waterworks.

The quality of drinking water of autonomous waterworks and individual intakes and wells is mostly unknown.

The paper tries to indicate the importance of quality control of any drinking water in the aim of population health protection.

It deals with the problems of fluoride content particularly in drinking water. This kind of groundwater quality problem in domestic expert and administrative society is not treated in right way.

It also tries to insight the problem of the origin of increased concentration of fluoride in some groundwater in the Srebrenica municipality and to point out serious

consequences of long-term consumption of the water with fluoride concentration higher than 1.2 mg/L.

1.1. Geographical and economical characteristics of the area

Srebrenica is a small city in the mountain area in the eastern part of the Republic of Srpska, Bosnia and Herzegovina (Figure 1). Total area of the municipality is 533.4 km². In accordance with the data of the Census 2013 there are 13409 inhabitants. About half of the population is settled in rural areas. The territory of the municipality is recognised from the ancient period as very important regarding metallic mineral resources, in the first order of lead and zinc, but also gold, silver, cadmium, arsenic, copper. The traces of mining reach in the Roman period. Lead, silver and gold were extracted from the ore in this period. Gaius Plinius Secundus (AD 23 – August 25, AD 79), better known as Pliny the Elder, mentioned in his papers the important production of gold in Pannonian and Dalmatian provinces. Related with these mineral resources there are also ferrous-arsenic mineral springs in Srebrenica vicinity. The most important one is "Crni Guber".



Figure 1: Srebrenica Municipality is located in the eastern part of the Republic of Srpska, Bosnia and Herzegovina

2. THEORETIC BACKGROUND

2.1. About fluoride and the concentration in groundwater in general

The presence of low or high concentrations of certain ions is the major issue that makes the groundwater unsuitable for various purposes. Fluoride is that kind of ions that causes health problems to people living in more than 25 nations around the world [4].

Fluoride is a ion of chemical element fluorite (atomic number 9) which belongs to the group of halogens (to 17th group or to VII group of the Periodic Table in accordance with earlier division).

It is a usual constituent of groundwater. Its concentration in groundwater is mostly less than 1 mg/L.

2.2. Fluoride Maximum Contaminant Level (mcl) in drinking water and consequences of concentrations above mcl

Fluoride concentration of at least 0.6 mg/L is required for human consumption because it makes stronger teeth and bones. In some countries with low concentration of fluoride in drinking water, like in Canada, it is usually enriched in fluoride before consumption [5].

From the other side, the consumption of water with fluoride concentration above 1.5 mg/L results in acute to chronic dental fluorosis where the teeth become coloured from yellow to brown.

Skeletal fluorosis, manifested by the weakness and bending of the bones, is also caused by the long-term consumption of water containing higher percentage of fluoride.

Precisely, the concentration of 3-10 mg/L causes skeletal fluorosis and long-term consumption of water with fluoride concentration more than 10 mg/L could result in more progressive type of fluorosis with significantly restricted mobility of consumer -crippling fluorosis.

Some other serious diseases caused by increased fluoride concentration in groundwater are: protoplasmic toxicant, impact on pineal gland, kidneys, urinary bladder, thyroid gland, gastro-intestinal tract etc.

The presence of high concentrations of fluoride in groundwater is natural or anthropogenic or a combination of both.

In accordance with domestic regulation, the Rule on Health Correctness of Drinking Water Quality,

maximum allowed concentration of fluoride in drinking water is up to 1.2 mg/L.

In comparison with the regulations of developed countries it is stricter.

For example, in Canada it is 1.5 mg/L [5].

The Environment Protection Agency (EPA) of the USA prescribes the Maximum Contaminant Level (MCL) up to 4 mg/L [6]. This level is considered as „health level“ (above this value of fluoride it is serious threat for human health).

The Secondary Maximum Contaminant Levels considered as so-called „aesthetic level“ and it is up to 2 mg/L. The concentration of fluoride in drinking water from 1 to 2 mg/L affects teeth colour but not seriously human health.

The concentration of 2-4 mg/L is „transitional zone“ between „aesthetic“ and „health“ level.

It is mentioned above that the concentration higher than 3 mg/L causes fluorosis and the concentration higher than 10 g/L causes the progressive type of this disease [6].

Recommendation of the World Health Organisation (WHO) [7] for fluoride concentration in drinking water is up to 1.5 mg/L.

2.3. Main reasons of increased concentration of fluoride in groundwater

There are two reasons of increased concentration of fluoride in groundwater – natural and anthropogenic.

The natural source of fluoride in groundwater is the presence of different types of the rocks and its interaction with groundwater.

As water flows through the rocks, it dissolves minerals listed in the table 1 and fluoride is naturally released into the water.

The natural concentration of fluoride in groundwater depends on the geological, chemical and physical characteristics of the aquifer, the porosity and acidity of the soil and rocks, the temperature, the action of other chemical elements, and the depth of the aquifer [8].

In accordance with the above-mentioned, fluoride content in groundwater could vary in wide range. Groundwater residence time is very important (the time of interaction between rocks and water).

For the condition of long-time interaction there is higher possibility for enriching of groundwater in fluoride, with prerequisite that groundwater flows through the rocks consist of minerals with fluorine.

The natural source of fluoride in groundwater includes different minerals in rocks, in the first order fluorite, apatite and micas [9].

The most important minerals are fluorite: CaF_2 , mostly emerged as the result of pneumatolytic processes, fluorite-apatite ($\text{Ca}_5(\text{PO}_4)_3\text{F}$) as well as micas (muscovite $\text{KAl}_2(\text{AlSi}_3\text{O}_{10})(\text{F},\text{OH})_2$ and biotite $\text{K}(\text{Mg},\text{Fe})_3\text{AlSi}_3\text{O}_{10}(\text{F},\text{OH})_2$) (Table 1).

Magmatic rocks have the content of fluorine from less than 100 ppm (ultramafite) to more than 1000 ppm (alkaline). Sedimentary rocks contain fluorine from 200 ppm (limestone) to 1000 ppm (schist) [10].

Groundwater with increased content of fluorine is mostly $\text{HCO}_3^- \text{Na}$, with relatively low content of Ca and Mg and pH value higher than 7.

The main minerals of fluorine, fluorite and sellaite, are not significantly soluble under normal pressure and temperature.

Their solubility rate increases in alkaline conditions and electro conductivity between 750 and 1750 $\mu\text{S}/\text{cm}$. Some explorations indicate that high content of fluoride in groundwater originates from biotite solubility [11].

Table1: Minerals containing fluorine

Mineral	Chemical content
Fluorite	CaF ₂
Sellaite	MgF ₂
Apatite	Ca ₅ (PO ₄) ₃ (F,Cl,OH)
Fluor-apatite	Ca ₅ (PO ₄) ₃ F
Cryolite	Na ₃ AlF ₆
Tremolite	Ca ₂ Mg ₅ Si ₈ O ₂₂ (OH) ₂
Hornblende	Ca ₂ (Mg,Fe,Al) ₅ (Al,Si) ₈ O ₂₂ (OH) ₂
Muscovite	KAl ₂ (AlSi ₃ O ₁₀)(F,OH) ₂
Biotite	K(Mg,Fe) ₃ AlSi ₃ O ₁₀ (F,OH) ₂
Sericite	KAl ₂ (AlSi ₃ O ₁₀)(F,OH) ₂

But generally considered, quantitative studies about fluoride in water, especially those treat equilibrium fluid-mineral are very rare.

The second reason of increased content of fluorite in groundwater is human activity. A number of human activities can also increase fluoride concentrations in groundwater including glass, steel and phosphate fertilizer production.

In addition, agricultural run-off, infiltration of fertilizers, and discharges from septic or sewage treatment facilities that process fluoridated water can all add inorganic fluorides to the environment.

The treated area is very sparsely populated and without any industrial activity (agriculture is minor).

It is the reason why natural conditions of high content of fluoride in some groundwater are considered as the most important.

3. EXPERIMENTAL PART

3.1. Samples collection

In the aim of the determination of drinking water quality in Srebrenica municipality rural areas more than 120 samples of groundwater were taken (Figure 2) from the representatives of the Institute for Water from Bijeljina [12]. The investigation is initiated by the local authorities with the aim of determination of main problem in groundwater quality within autonomous and non-centralised waterworks.

The sampling had been performed in the period between June 18th and July 19th 2010. Here are 103 samples considered in this paper. For the rest of the samples it was not possible to provide the exact location based on the available sheet records on the sampling point.

The samples were taken and conserved in accordance to the BAS ISO 5667-2, 3 and 11 standards [13,14]. The parameters like temperature, pH and electro conductivity are measured insitu, odour and taste as well.

The review of the sampling points per local communities is visible in the figure 3.

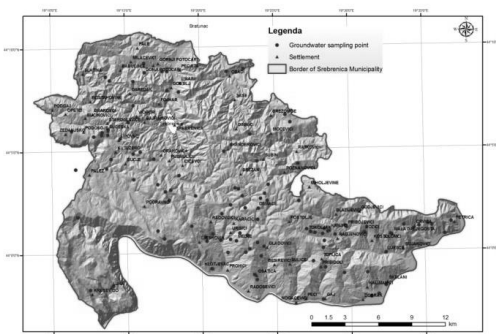


Figure 2: Groundwater sampling points

3.2. Laboratory analysis

The analyses were performed in the laboratory of the Institute for Water from Bijeljina during the summer 2010. Fluoride is determined in ionic chromatograph in accordance with BAS EN ISO10304-1:2010.

This method is applicable for surface water and groundwater as well as drinking water. Liquid chromatography ionic separation took place on chromatograph column. Anionic exchanger of low capacity is used as stationary phase and liquid solution of salt of low monobasic or dibasic acid as mobile phase.

Fluoride detection is based on electro conductivity. For the anion detection the suppressor which decrease electro conductivity of eluent is applied.

In suppressor, separated anions are converted in their acid forms of high conductivity. Carbonate-bicarbonate eluent is converted in carbonate acid of low conductivity.

Separated anions, in their acid forms, are measured based on conductivity. They are identified in accordance with retention time. Quantitative analysis is based on the high of the pick or area beneath the pick.

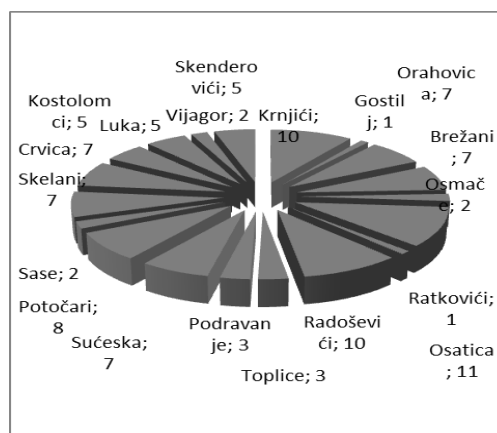


Figure3: Number of samples per local community during the campaign June-July 2010

4. THE RESULTS AND DISCUSSION

4.1. The reasons of increased content of fluoride in some groundwater of Srebrnica municipality

The background for analysis of the fluoride concentration in drinking water is the Rule on Health Correctness of Drinking Water Quality. Additionally, some other standards are considered, in the first order the standards of the World Health Organization and the USA Environment Protection Agency (EPA). Detail explanation of these standards is given in the chapter 4.

Taking into account the absence of industrial and agriculture activities in the area of interest it is concluded that the cause of high concentration of fluoride in some groundwater is natural or precisely geological (aftermath of the interaction between groundwater and rocks during the groundwater flow in the aquifer).

Further, geological conditions are considered on the base of the data presented in the Basic Geological Map 1:100000, the sheets Ljubovija, Višegrad, Valjevo and TitovoUžice [15].

Simplified geological map of the area of interest is shown in the Figure 5. It is obvious that three groups of the rocks of different age and lithology prevail:

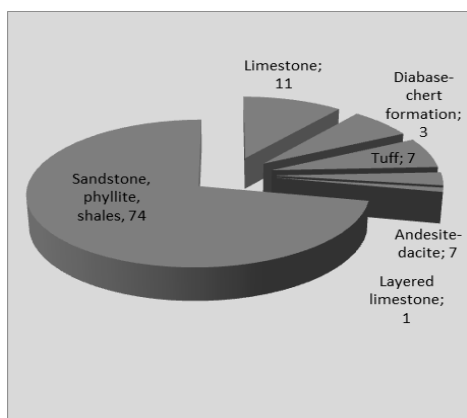


Figure 4: Groundwater samples number per litho-stratigraphic units

- Quaternary sediments – debris (s), alluvial (al) and deluvial sediments (d)
- Tertiary (Oligocene and Miocene) - conglomerates and clays (Ol,M) and the Miocene volcanic (andesite and dacite) and pyroclastic (tuffs) rocks, the result of the Srebrenica Tertiary volcanism (□ and □Tc);
- Mesozoic: Triassic sandstones (T1), limestones (T2) and subordinately diabase (□□); Jurassic (J2,3) diabase-chert formation; Cretaceous layered limestones (K2)
- Paleozoic sandstones, phyllites and schists (1C1,2;2C1,2;3C1,2).

Consideration of the rocks and spatial fluoride anomalies distribution in groundwater are the keys for the problem determination. In total, 103 samples were taken from 6 different litho-stratigraphic units (Table 2) and they are considered as rather enough for this kind of analysis.

The most samples were taken from the springs that occur in three packages of sedimentary and metamorphic rocks of the Lower-Mid Carboniferous age (1-3C1,2) (Figure 5). Groundwater samples number per lithological units and percentage share of those with $F > 1.2$ mg/L are visible in the Table 2.

It is obvious from the table that excessive concentration of fluoride exclusively occurs in the aquifers created in the sediments of the Carboniferous age, related to their mineralogical content (Figure 4).

Table 2: Samples number per litho-stratigraphic units and percentage of those with $F > 1.2$ mg/L

Lithology	Age	Samples number	Samples number with $F > 1.2$ mg/L	% samples with $F > 1.2$ mg/L
Limestone	T ₂ , T _{2,3}	11	0	0.00
Andesite-dacite	a	7	0	0.00
Tuff	Tc	7	0	0.00
Diabase-chert formation	J _{2,3}	3	0	0.00
Layered limestone	K ₂	1	0	0.00
Sandstone, phyllite, schists	C _{1,2}	74	44	59.42

It is important to emphasize that geological explorations in the past were not detected localities with high content of fluoride and apatite in Srebrenica Municipality. On the other hand, a significant content of mica minerals in the Carboniferous sandstones is well-known [15]. It is the reason why mica minerals (in the first order muscovite and biotite, Figure 6) are recognised as the key minerals responsible for higher content of fluoride than usual (mineral formula of biotite is given in the Table 1) and why fluoride exclusively exceed the allowed limit (1.2 mg/L) in samples from the Carboniferous sandstones.



Figure 5: Simplified geological map of Srebrenica area with groundwater sampling points

From 74 samples from the Carboniferous sandstone and schists even 44 or 59.42 % exceed maximum allowed concentration in drinking water in accordance with the actual legal policy. Thirty six samples or 48.64% exceed the Secondary Maximum Contaminant Level or so-called "aesthetic standard" of the EPA ($F > 2$ mg/L), and in 2 samples F concentration exceeds 10 mg/L.

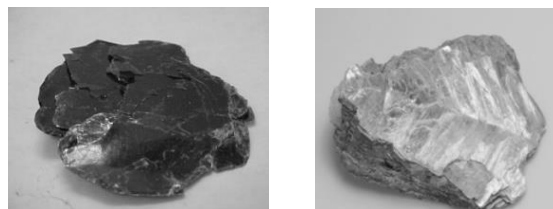


Figure 6: Trioctahedral micas biotite and dioctahedral muscovite

It is probably the result of the solubility of the mica minerals in the Carboniferous rocks.

In the other rocks there are no samples in which fluoride exceeds maximum allowed concentration for drinking water. It is necessary to emphasize that most important fluorine minerals are constituents of ore veins, especially lead-zinc veins. It means that increased concentration of fluoride should be expected in andesite-dacite rocks close to lead-zinc veins. From the Table 2 it is obvious that there are no samples from andesite-dacite rocks with increased fluoride concentration. The reason probably lies in the fact that taken samples are distanced from the ore veins. However, the groundwater from volcanic rocks, especially those close to the lead-zinc veins (e.g. in Sase area) should be analysed in detail in the future.

4.2 The outputs of the fluoride content analyses

From the previous analysis increased content of fluoride in groundwater in Srebrenica Municipality is the result of the interaction between groundwater and rocks that contain mica minerals (muscovite, biotite, sericite).

The areas with fluoride content above 1.2 mg/L (above allowed value for drinking water in accordance with the Rule) are visible in the Figure 7.

Table3: Minerals with F which constitute the rocks of the Carboniferous age ($^{1-3}C_{1,2}$) in the territory of Srebrenica municipality

Rock	Age	Mineral	Chemical content
Phyllite	$^1C_{1,2}$	Muscovite	$KAl_2(AlSi_3O_{10})(F,OH)_2$
		Muscovite	$KAl_2(AlSi_3O_{10})(F,OH)_2$
Meta-sandstone	$^2C_{1,2}$	Muscovite	$KAl_2(AlSi_3O_{10})(F,OH)_2$
Meta-sandstone		Muscovite	$KAl_2(AlSi_3O_{10})(F,OH)_2$
Sericite schist	$^3C_{1,2}$	Sericite	$KAl_2(AlSi_3O_{10})(F,OH)_2$
Meta-sandstone		Muscovite	$KAl_2(AlSi_3O_{10})(F,OH)_2$
Sandstone	$^3C_{1,2}$	Biotite	$K(Mg,Fe)_3AlSi_3O_{10}(F,OH)_2$
		Sericite	$KAl_2(AlSi_3O_{10})(F,OH)_2$

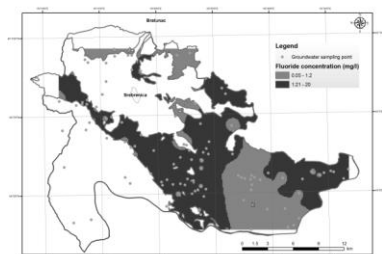


Figure 7: Spatial distribution of the zones with fluoride content lower and higher than 1.2 mg/L in the Carboniferous sediments of Srebrenica Municipality

In the figure 8 isolines of F ion concentration in groundwater are shown. Of course, the distribution of F ion in groundwater must be considered with all limitations arises from the samples number and interpolation method (Inverse Distance Weight).

In accordance with spatial distribution of fluoride in groundwater (Figure 8) it is possible to define two greater and three smaller zones of fluoride anomalies in groundwater (Figure 9).

The greatest anomaly zone extends from the central part of the municipality (Subin, Brežani, Osmače) up to the overall southern part of the municipality (Ljeskovik, Radoševići) (Figure 9). East from this zone high increment in fluoride concentration is found in the area of the villages: Bučinovići, Opetci, Sućeska, Bektići, Lipovac, Kutuzero, Pasmulići, Orahovica and Jasenova.

The zone extended from Kostolomci via Crvica and Daljegošta up to Petrica is particularly interesting. Registered fluoride concentrations in 6 springs of this zone are higher than 3.5 mg/L.

The maximum values in the territory of Srebrenica Municipality is recorded right here. It is registered in the spring "Edina voda" in Daljegošta village, 16.11 mg/L. The next one is registered in Crvica village, 12.88 mg/L. It is probably effected by very high concentration of micas in the rocks Carboniferous age.

One smaller zone of fluoride anomalies in groundwater is registered between Fojhar and Gostilj villages (Figure 9).

It is realistic assumption about increased content of mica minerals in the Carboniferous sandstones and schists in these areas.

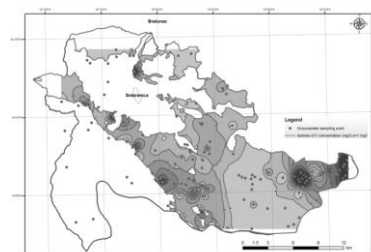


Figure 8: Isolines of groundwater F ion concentration in the territory of Srebrenica Municipality

4.3. The outputs of possible impacts of increased fluoride concentration in groundwater on health

Even 36 samples have fluoride concentration above 2 mg/L, 24 above 3 mg/L and 9 above 6 mg/L. Two samples have the concentration higher than 10 mg/L. Taking into account the EPA standards, potential health impacts are given in Table 4. For further analyses of the impact of fluoride concentration in drinking water on population, it is necessary to collect and process the data on consumers' history of diseases, especially those related to skeletal and dental diseases in the areas with fluoride concentration in groundwater higher than 2 mg/L. It is especially interesting to collect the data on skeletal diseases of the population in the area Daljegošta-Crvica-Petrica, where 5 of 6 taken groundwater samples have fluoride concentration higher than 5 mg/L.

The concentrations in Daljegošta and Crvica are 10.7 and 8.6 times higher than concentration allowed by domestic legal policy.

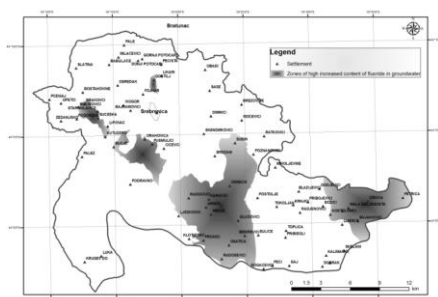


Figure 9: Zones of fluoride content in groundwater higher than 3 mg/L



Figure 10: Areas of possible increased values of fluoride (>1.2 mg/L) in groundwater in the territory of the Republic of Srpska (red polygons)

Table 4: Potential health impact related with fluoride content in groundwater

Health impact	F ⁻ content (mg/L)	No of GW samples	Settlement with possible impact
Dental fluorosis	>2	36	Brežani, Osmaće, Murselovići, Karačići, Kiprovo, Opetci as well as each settlement listed bellow
Skeletal fluorosis	3-10	24	Orahovica, Podosmaće, Gladovići, Karno, Osatica, Radoševac, Urisići, Ljeskovik, Jasenovca, Bektić, Sućeska, Brđani, Crvica, Daljjeogošta, Subin
Crippling fluorosis	>10	2	Daljjeogošta, Crvica

4.4. Application of the results for the rest of the territory of the Republic of Srpska

Groundwater analyses in the mentioned zones should provide a clear picture of the impact of the rocks of the Carboniferous age and its mineralogical content on the creation of chemical composition of groundwater and the relation with fluoride concentration. It is also important to continue sampling and analyses of groundwater in Srebrenica area, but also in the sandstones, phyllite and schist of the Middle and the Upper Carboniferous in the territories of the municipalities Milići and Bratunac.

In analogy with presented results, it is very important to collect data about groundwater quality within the rocks of the Carboniferous age in the rest of the territory of the Republic of Srpska. In the first order there is Una-Sana Paleozoic in the west, but also Prača Paleozoic in the south-east part of the Republic of Srpska (Figure 10).

5. CONCLUSION

The presence of low or high concentrations of certain ions is the major issue that makes the groundwater unsuitable for various purposes. Fluoride, as usual constituent of groundwater in concentration mostly less than 1 mg/L, is kind of ion that causes health problems to people living in more than 25 nations around the world. Fluoride concentration of at least 0.6 mg/L is required for human consumption. In contrast, the consumption of water with fluoride concentration above 1.5 mg/L results in acute to chronic dental, even skeletal fluorosis. In accordance with domestic standards maximum allowed concentration of fluoride in drinking water is up to 1.2 mg/L.

In the aim of the determination of drinking water quality in Srebrenica municipality rural areas more than 100 samples of groundwater were taken. The most samples were taken from the springs that occur in three packages of sedimentary-metamorphic complex of the Lower-Mid Carboniferous age.

From 74 groundwater samples from the Carboniferous sandstones, phyllites and schists even 44 or 59.42 % exceed maximum allowed concentration in drinking water in accordance with the actual standards. In 2 samples F concentration exceeds even 10 mg/L. It is realistic assumption about increased content of fluoride in groundwater because mica minerals in the Carboniferous sedimentary-metamorphic complex.

The greatest anomaly zone extends from the central part of the municipality up to the overall southern part of the municipality. East from this zone high increment in fluoride concentration is observed in villages: Bučinovići, Opetci, Sućeska, Bektići, Lipovac, Kutuzero, Pustumlići, Orahovica and Jasenovca. The zone extended from Kostolomci via Crvica and Daljjeogošta up to Petrica is particularly interesting. Registered fluoride concentrations in 6 springs of this zone are higher than 3.5 mg/L. The maximum values is recorded right there. It is registered in the spring "Edina voda" in Daljjeogošta village, 16.11 mg/L. The next one is registered in Crvica village, 12.88 mg/L.

For further analyses of the impact of high fluoride concentration in drinking water on human health it is necessary to collect and process the data on consumers' history of diseases, especially those related to skeletal and dental diseases in the areas with fluoride concentration in groundwater higher than 2 mg/L. It should be investigated as soon as possible especially in the area Daljjeogošta-Crvica-Petriča.

Based on the above-mentioned facts it is very important to collect data about groundwater quality within the rocks of the Carboniferous age in the rest of the territory of the Republic of Srpska. In the first order there is Una-Sana Paleozoic in the west, but also Prača Paleozoic in the south-east part of the Republic of Srpska.

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