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Investigating Recycled Filter Media in order to Remove Fluoride Compounds from Groundwater

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Abstract. In this article, an enhanced recycled filter media for the removal of fluoride compounds from groundwater via filtration process was investigated. Experiments were made to investigate the influence of recycled filter media on removal efficiency of fluoride compounds. The recycled backwash cake based filter media used at Lithuanian water works were investigated in a pilot test-bench constructed for this research in the Water Management Laboratory of Vilnius Gediminas Technical University.

Keywords: backwash residual, groundwater, fluoride removal, filtration.

Conference topic: Water engineering.

Introduction

Drinking water supplied to Kretinga town (Lithuania) from eastern well field Padvariu, which exploited two water horizons, i.e. upper Permian and upper Devon Žagarė, water level that is situated at 40–50 m deep and with a highest water horizon, which is situated deeper at 90–103 m.

The waterworks started working in last decade. From its beginning the waterworks used approximately 100 m^3/d water. The water needs of Kretinga town became lower from installation was started to use and only approximately 18 000 inhabitants were used water last year: from Padvariai, Salantai, Vydmantai, Darbėnai and Kūlupėnai well fields.

The town everyday needs are approximately estimated to be lower need of water and the future needs will be approximately 30% lower. Hydro-geological investigation's data show that such a quantity of water can ensure well field Padvarių water horizon alone or with upper Permian and upper Devon Žagarė horizon water level.

Water resources of the upper Permian and upper Devon Žagarė are sufficient in quantity, also the water horizons of Kretinga region are hydraulically connected and because of this if exploitation of upper Permian and upper Devon Žagarė will be more intensive, the water quality of one third of the waterworks can became lower.

The quality of different groundwater horizons is not the same. The quality of groundwater from upper Permian and upper Devon Žagarė horizon following "Kretingos vandenys" data during 2014–2015 is: turbidity 0.25–0.71 mg/L, colour 2.5–7.5 mg/L Pt, total iron concentration – 1.7–1.1 mg/L, manganese – 0.02–0.01 mg/L, ammonia – 0.31–0.18 mg/L, Chemical Oxygen Demand (COD) – 0.80 mgO_2/L, fluoride concentration – 1.7–2.1 mg/L. Water quality from the Vydmantu water field- turbidity till 1.2 mg/L, colour – till 5 mg/L Pt, total iron concentration – till 0.15 mg/L, manganese – equal or less 0.01 mg/L, ammonia – till 0.43 mg/L, COD – till 1.5 mgO_2/L, fluoride concentration – till 2.8 mg/L.

Groundwater is supplied to the drinking water treatment system with fluoride removal equipment, after that the water is disinfected and supplied to the town networks. Fluoride removal plants were built following the typically module design with simple aeration of groundwater and filtration via reverse osmosis membranes with rejected by membranes concentrated water (till 5 mg/L fluoride concentration and approximately 30% of total prepared water) membrane filtration technology. Fluoride concentration of filtered water is 0.1 mg/L and sometimes less. Prepared water conductivity decreases a little. Kretinga town drinking water from the centralised water supply has a quality following both European Directive and Lithuanian Hygiene Norm. The reason for the negative social reaction and acceptance of the water from the centralised water supply is the high tariff of drinking water in the town, indicating a high level of water preparation cost. Social problems are connected with payments by using one of the highest in Lithuania tariff, during the highest use of membrane operated water treatment works some high electricity power use can be occurred and that can have some influence on the negative reaction of the supplied water. The last conclusions made have a strong argument, because the high level of tariff existing in the town, which cannot be changed by using membrane filtration, and after the payments estimating can form social and economic misbalance. Kretinga town water works and the existing water treatment equipment experience following the conclusions of hydrogeology and health specialists about the negative fluoride influence on water user's health conditions (Wasana *et al.* 2016) of the drinking water after fluoride removal shows that the fluoride content in the groundwater (Jadhav *et al.* 2015) is completely or partially can be removed in a complex of filtration using iron enriched compounds form (Sujana *et al.* 2009). Fluoride removal connected with iron hydroxide compounds needs to follow the sorption and filtration processes (Tang *et al.* 2009). Iron, fluoride, water pH and other compounds (ions presented in groundwater) concentrations need to be decreased if the quality of treated water should follow a required quality classification (Valentukeviciene 2009). The problem with unacceptable fluoride concentration can be solved in the simple filtration (Reardon, Wang 2000; Smittakorn *et al.* 2010). The existing water treatment at Kretinga can be used for the removal of fluoride using conventional filtration.

Methods and materials

Two technologies (Castel *et al.* 2000) can be proposed for the water works water treatment allowing for required quality drinking water classification, except for membrane technologies (Jadhav *et al.* 2014), because it needs a lot of energy:

- technologies with chemicals: using strong acids, coagulation and filtration (Chung *et al.* 2015);
- Technologies without chemicals: using of adsorbents and filtration (Oladoja *et al.* 2016; Ramdani *et al.* 2010; Xu *et al.* 2015).

Most common adsorbent material for the fluoride removal is aluminium hydroxide (granulated or filter media), but it is artificial material and can negatively influence the water's quality. Aluminium hydroxide is quite expensive compared with adsorption effectiveness (Tomar, Kumar 2013). Some kinds of recycled filter media that are excavated in Lithuania and imported from the Poland can be used for the water treatment at fluoride removal water works. The investigation's results have shown the effectiveness of iron enriched filters backwash to adsorb fluoride compounds.

Aim of research – experimental investigation of water treatment technology without chemicals, when groundwater is treated by a recycled adsorbent (granulated filter backwash) and filtration is done with iron enriched filter media at the laboratory scaled filtration column.

A pilot scale experimental plant was built at Vilnius Gediminas Technical University in laboratory; the principal scheme is shown in Figure 1. Obtained groundwater from the main pipeline was supplied by a pump to the water supply's pipeline, connected to the reservoir and pilot scale filters Nr. 1 and Nr. 2. The construction of the reservoir leads to the regular contact time (by volume changing) depending on the technological needs. Recycled filter media was added to the filtration column. All contents of the reservoir volume were mixed by an additive mixing without the dosage of conventional chemicals. Groundwater and aerated flow mixture from the reservoir was supplied to the filtration equipment during the sorption process of the recycled filter media. Filtration equipment was constructed from two DN50, 1500 mm lengths of plastic columns. Water after filtration was supplied to the pilot scale reservoir Nr. 2 during the sampling of fluoride and other compounds. All pilot scale filter construction is the same before and after regeneration procedure and presented in Figure 1. Layers high and the particle sizes were the same, but filter media were regenerated using 10% solution of NaOH. Filter media of pilot the scale filter Nr. 1 was made from new iron enriched backwash material and filter Nr. 2 – made from used for fluoride removal regenerated filter media (fresh filter media is ready for fluoride removal after backwash treatment and for continues fluoride removal – after regeneration). Experimental equipment lead compares treatment by adsorbent and untreated aerated filtration via iron enriched filter media filtration technology, when the same groundwater water was supplied to the reservoir and after aeration to the pilot scale filters Nr. 1 and Nr. 2. Pressure losses were metered with piezometers. The filter media was backwashed with water flow from centralised water supply. Special taps regulated the experimental pilot scale plant; equipped in the untreated water supply pipelines and the filtrated water outlet pipeline and the water flow was metered by volume method.

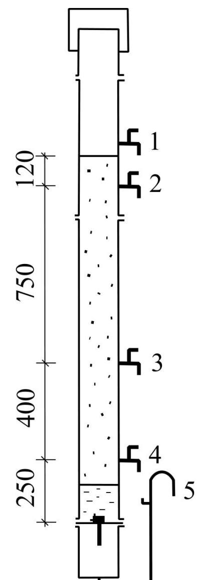

Fig. 1. Pilot scale plant consists of: 1 – taps for the groundwater analyses taking; 2 – taps for the inlet-water analyses taking; 3 – taps for the internal water analyses taking; 4 – taps for the filtered water analyses taking; 5 – taps for the prepared water analyses taking

Merck systems Spectrophotometer were used for the fast determination of fluoride and other compounds concentrations. The water conductivity, pH parameter, colour and turbidity were determinate by standard methods.

The dosage rate 5 g/L was appropriated by investigation results from the laboratory's research before the experiment when the water and adsorbent contact time was 25 min. The water flow rate was 3 m/h at filtration equipment (facilities of filtration equipment was approximately equal to 12 m^3/m^2 h). Groundwater was supplied at the same time to the all-pilot scale filters (to the filtration columns – via a contact reservoir and the filtration equipment). All pilot scale filters and filter media were backwashed properly with water supplied from centralaside water supply before every filter rate until the backwashing water seemed clear. The filter rate was established and filter pressure losses were registered before every start of the filtration rate. The filtration rate was controlled, filter media pressure losses were registered, and an analysis of the untreated water, water after filtration equipment and filtrated water were taken (depending on filter media layers) after 25 min filtration time (when the first filtrated water occurred). Untreated water analyses were taken twice during the filtration time and another analyses every 2–8 hours. Filter media pressure losses were registered all the time and the filter rate was controlled by the situation. The water supply was stopped when the filter media pressure losses were raised to 2 m w. t. and at the end the filtration rate was registered and the backwashing was started. Investigation of filter media microstructure was carried out by employing SEM "Quanta" 250 with SE detector.

Results and discussions

Filter media consists of spherical particles of various sizes. Size of these particles is approximately 150–350 µm. In addition, fragments of these particles are seen can be seen from Figure 2. Large amount of hollow areas exists between the connected individual particles. Surface of the particles is granular, formed from many small particles that are connected in one conglamerate. Between these particles open, small and interconnected pores exist for the efficient sorption process.

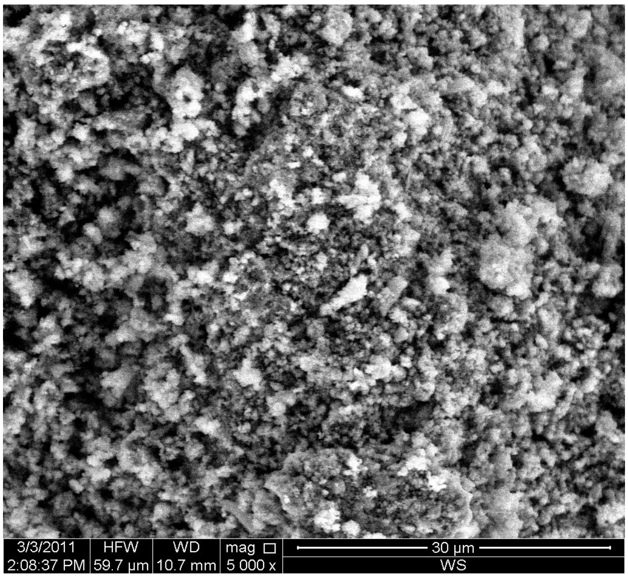

Fig. 2. SEM photo of sludge microstructure particle (5.000×)

It was identified that bulk density of backwash residual particles is 547 kg/m^3, particles' density 2270 kg/m^3, specific surface area 9657 cm^2/g.

Chemical analysis of iron enriched filter backwash residual shows that 68% of obtained dry residual consists of Fe_2O_3, 10% of SiO_2, 8% CaO as well as 9% of P_2O_5 exist. The amounts of other oxides do not exceed 3%. It was identified that pH of filter backwash residual is 7.81, i.e. slightly alkaline medium.

The biggest part of the experiment was done with groundwater from the upper Permian and upper Devon Žagarės horizons. Data of the experimental investigation was statistically calculated from the registered analyses and are presented in figures below.

Filtrated water quality results of fluoride concentration, conductivity and pH indicator following European Directive and Lithuanian Hygiene Norm for acceptable quality of drinking water category resulted if the filtration rate is 5 m/h and fluoride concentration, pH index and colour of treated water are not high. Only the undetectable part of fluoride lower than 0.1 mg/L still found in filtrated water (fluoride concentration in untreated water was up to 2.4 mg/L); and the pH index decrease to 7.4. The time of filtration rate was long, and pressure losses after 8 hours were only 1.64 m w. t., because the pilot scale filter media removed only a small part of the pollutants (relatively low amounts of other compounds were in untreated water). The same quality water was filtered in the same conditions by pilot scale filter Nr. 2 with satisfactory results. The reason for the satisfactory results is because the filter media of the pilot scale filter Nr. 2 was of new filter media and the filter media of the pilot scale filter Nr. 1was of regenerated filter media.

During the upper Permian and upper Devon Žagarės horizon groundwater treatment with aeration and filtration when the filtration rate is 7 m/h and the filter media is 0.6–2.5 mm fraction size, 1.5 m high iron enriched filter media, results of the drinking water's quality following the acceptable quality category. Other part of the investigation was done with a slygtely different quality upper Permian and upper Devon Žagarės horizon groundwater treatment.

The following results during the aeration and filtration rate were: 1–3 m/h of untreated groundwater with fluoride concentration 2.757 mg/L, pH index 7.44–7.62, and water conductivity from 600 μScm^{-1} (25°C), via pilot scaled filters Nr. 1 and Nr. 2, fluoride removal was 80–86% (concentration in filtrated water was less than 1.5 mg/L), pH indexes variation was only 22–35% (pH in filtrated water till 7.62). Variation of conductivity was measured too (variation was approximately 50%), and other indexes stay without decreasing. Filtration rate time was 24–30 hours before pressure losses increased up to 2 m w. t.

The following results during the aeration, 20–25 min. adsorption with iron enriched adsorbent and filtration rate were: 5 m/h via pilot scaled filter Nr. 2 filter media of Kretinga town groundwater, concentration of the fluoride decreased to 1.5 mg/L, pH index-to 7.60–7.64, water conductivity decresed below 300 μScm^{-1} (25°C), water colour indicator was measured till 15 mg/L Pt. Fluoride concentration of the filtered water was more than 1.20 mg/L. Water quality increased during the groundwater treatment with iron enriched adsorbent: fluoride concentration efficiently decrease from 2.70 till 1.50 mg/L, water conductivity variation – 50–60% (from 600 till 300 μScm^{-1} (25°C), pH index decrease from 8.23 till 7.44. Concentration of fluoride in supplied groundwater decreases a little – from 2.70 to 2.60 mg/L. The fluoride removal effectiveness was satisfactory following the accepted conditions and the water treatment parameters were changed: dosage of 10% NaOH solution for the regeneration was increased up to 80–90 g/L, filtration rate was decreased to 3 m/h. Concentrations of fluoride before and after filter media regeneration and conductivity decrease can be seen in the last filtration rate data showed in Figures 3, 4 and 5.

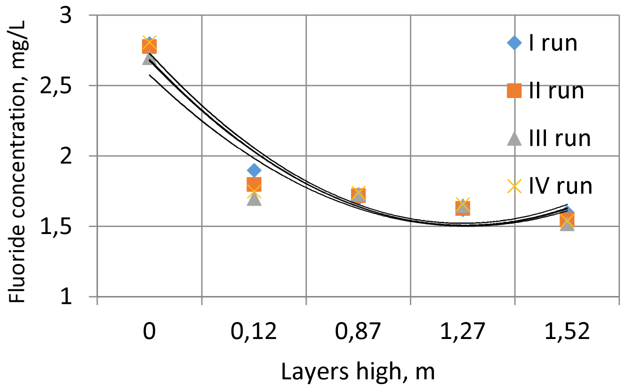

Fig. 3. Fluoride removal from the Kretinga town groundwater filtered 5 m/h filtration rate, when groundwater content $F^- = 2.75$ mg/L, water after filtration $F^- = 1.50$ mg/l (pilot scaled filter Nr. 1)

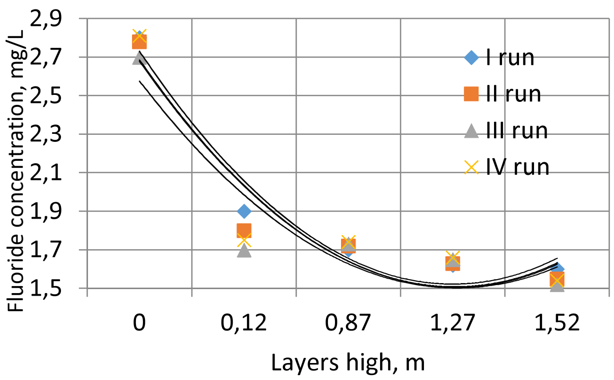

Fig. 4. Fluoride removal using regenerated filter media, groundwater filtered 3 m/h filtration rate, when groundwater content was $F^- = 2.75$ mg/L, water after filtration – $F^- = 1.50$ mg/L (pilot scaled filter Nr. 2)

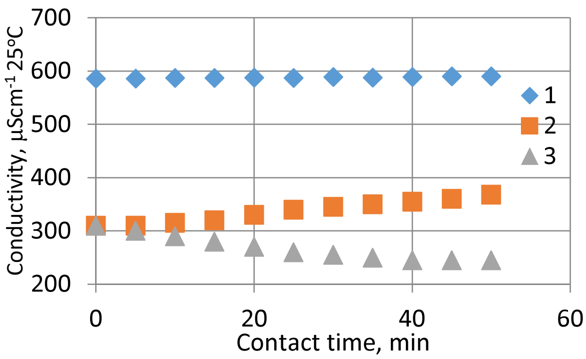

Fig. 5. Changes of the conductivity for the treated groundwater when treated with iron enriched sorbent and filtrated 5 m/h rate (pilot scaled filters Nr. 1 and 2, 1 – groundwater, 2 – water after filtration treatment, 3 – filtered water after filter media regeneration)

Conclusions

1. When groundwater of Kretinga town (Lithuania) from eastern well field Padvarių, which exploited two water horizons, i.e. upper Permian and upper Devon Žagarė was aerated and filtrated with a filtration rate of 5 m/h via 0.65–2.5 mm particle size filter media, 1.52 m high iron enriched backwash residual, the filtered water was of acceptable quality drinking water category.
2. Dosage of 10% NaOH solution into the filter media for regeneration purpose has positive influences on the treated water's quality. Dosage of 80 g/L 10% NaOH, when contact time is 25 min., after filtration treatment of the fluoride concentration of water decrease from 2.75 to 1.50 mg/l, manganese – 50–60% (from 0.20–0.18 to 0.08–0.09), pH index decrease from 8.23 till 7.44. Water conductivity variation 50–60% was in acceptable following European Directive and Lithuanian Hygiene Norm – from 600 till 300 μScm^{-1} (25°C).
3. When groundwater of groundwater of Kretinga town (Lithuania) water works was aerated, treated with iron enriched recycled backwash residual and filtrated with filtration rate 3 m/h via 0.65–2.5 mm particle size filter media, 1.52 m high filter media (fresh and regenerated), filtered water fluoride concentration was below 1.5 mg/L (untreated water till 2.75 mg/L), pH index till 8.23, water conductivity was in acceptable limits. Water pH index was the same after filtration. All results of fluoride concentration after treatment shows that iron enriched backwash residual is an in a stable sorbent cand without membrane filtration or strong chemical compounds it cannot be usefull and efficient for fluoride removal from groundwater.
4. When groundwater of groundwater of Kretinga town (Lithuania) water works was treated with adsorbent enriched with iron compounds, water conductivity decreased more than by half. Without treatment with adsorbent groundwater conductivity there is a negative influence on the water related equipment and interfere with sedimentation on internal surface.
5. When the dosage of regenerating solution 10% NaOH was increased up to 80–90 g/L and filtration rate decreased to 3 m/h, groundwater of groundwater of Kretinga town (Lithuania) water works was aerated, treated with iron enriched adsorbent, using sedimentation equipment filtrated via 0.65–2.5 mm particle size filter media, 1.52 m high backwash residual), filtrated water fluoride concentration was approximately 1.5 mg/L, water conductivity approximately till 300 μScm^{-1} (25°C); all the water's quality was of a required quality drinking water category.

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